



## **The radiological exposure of man from ingestion of Cs-137 and Sr-90 in seafood from the Baltic Sea. Pilot project: Marina-Balt**

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*Publication date:*  
1995

*Document Version*  
Publisher's PDF, also known as Version of record

[Link back to DTU Orbit](#)

*Citation (APA):*  
Nielsen, S. P., Øhlenschläger, M., & Karlberg, O. (1995). *The radiological exposure of man from ingestion of Cs-137 and Sr-90 in seafood from the Baltic Sea. Pilot project: Marina-Balt*. Risø National Laboratory. Denmark. Forskningscenter Risoe. Risoe-R No. 819(EN)

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# **The Radiological Exposure of Man from Ingestion of Cs-137 and Sr-90 in Seafood from the Baltic Sea**

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**Risø-R-819(EN)**

**Pilot Project: Marina-Balt**

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**Abstract** This report describes a limited radiological assessment of the collective doses to man from the intake of seafood from the Baltic Sea contaminated with the radionuclides Cs-137 and Sr-90. Information on fisheries statistics is presented. The most important source terms to radioactive contamination of Cs-137 and Sr-90 in the Baltic Sea are identified and quantified. A compartment model for the dispersion of radionuclides in European coastal waters including the Baltic Sea is described and tested by comparing model predictions with observations. Collective doses are calculated with the model for each of the source-term categories.

ISBN 87-550-2072-0

ISSN 0106-2840

Grafisk Service, Risø, 1995

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# 1 Introduction

The Baltic Sea is a semi-enclosed and shallow sea in which dissolved substances remain for a relatively long period of time compared to many other areas. This leads to greater accumulation of pollutants in seawater and in sediments than in other areas of the Atlantic Basin. Pollutants are transferred to man via marine pathways, i.e. consumption of fish of which there is an annual harvest from the Baltic Sea of about 600,000 tonnes.

This report describes a limited radiological assessment of the collective doses to man from the intake of seafood from the Baltic Sea contaminated with the radionuclides Cs-137 and Sr-90. Information on fisheries statistics is presented. The most important source terms to radioactive contamination of Cs-137 and Sr-90 in the Baltic Sea are identified and quantified. A compartment model for the dispersion of radionuclides in European coastal waters including the Baltic Sea is described and tested by comparing model predictions with observations. Collective doses are calculated with the model for each of the source-term categories.

This work is done for the Commission of the European Communities, Directorate General XI, Environment, Nuclear Safety and Civil Protection under Contract No. 94-PR-022.

## 2 Fisheries Statistics

The fisheries statistics have been obtained from the International Council for the Explorations of the Seas (ICES 1993) from 1991 and are reproduced in Table 2.1. The table shows the catch for different regions of the Baltic Sea. The fish catch is dominated by the species herring (60%), cod (20%), sprat (16%), and the remainder concerns flounder, salmon, dab, trout, turbot and plaice.

## 3 Source Terms

The following four source terms have been considered: nuclear weapons fallout, Chernobyl fallout, liquid discharges from the European reprocessing facilities Sellafield and La Hague, and liquid discharges from nuclear installations bordering the Baltic Sea.

Fallout of Cs-137 and Sr-90 from the atmospheric nuclear tests has been based on measurements made in Denmark. These data are considered representative for the Baltic Sea area. The data cover the period from 1955 to 1992 (Aarkrog et al. 1992) as shown in Figure 3.1.

The catchment area of the Baltic Sea ( $1.7 \cdot 10^{12} \text{ m}^2$ ) is about four times the water area ( $4.0 \cdot 10^{11} \text{ m}^2$ ), and the transfer of terrestrial fallout with river water to the sea is an important contribution to consider. Salo et al. (1985) investigated the inventories of Cs-137 and Sr-90 in seawater and sediments of the Baltic Sea for the time period 1961 to 1981 and give data for the total runoff of Cs-137 and Sr-90 with the rivers. Their data show that the river runoff of Sr-90 for the period 1961-1981 constitutes an important contribution to the input of Sr-90 into the Baltic seawater and since 1972 the river runoff has dominated over the direct

fallout from the atmosphere to the water. For Cs-137 the river runoff is less important - of the order of a few per cent compared to the atmospheric fallout into the water. We have constructed two simple sub-models and fitted them to these data allowing extrapolations to be made for the runoff of Cs-137 and Sr-90 into the Baltic seawater. The predictions and the data are shown in Figures 3.2 and 3.3 which give for the time period 1950 to 2000 the estimated input of Cs-137 and Sr-90 via rivers to the Baltic seawater. The sub-models are based on the classical UNSCEAR technique (eg. UNSCEAR 1972) using multiple regression of the data from 1961 to 1981 against the annual fallout rates and the accumulated fallout concentrations. The following sub-models were determined:

$$\text{Sr-90: } R_i = 13 d_{i-1} + 14 d_{i-2} + 3 A_{2H} + 8 A_i$$

$$\text{Cs-137: } R_i = 1.1 d_i + 3.4 d_{i-1} + 1.1 A_i$$

The total river runoff  $R_i$  (TBq a<sup>-1</sup>) in year  $i$  is a function of the fallout rate  $d_i$  (Bq m<sup>-2</sup> a<sup>-1</sup>) in the same year, and/or the previous years  $d_{i-1}$ ,  $d_{i-2}$ , and the accumulated fallout concentrations  $A_{iH}$  (Bq m<sup>-2</sup>), where  $H$  designates the effective halflife. Both regressions give a  $R^2$ -value (coefficient of determination) of 0.97. The two sub-models illustrate the radioecological characteristics for the transfer from soil (to vegetation) of Sr-90 and Cs-137. The transfer of Sr-90 is influenced by a time lag of one to two years and the accumulated fallout based on the physical halflife and a much shorter halflife. The transfer of Cs-137 depends on the recent fallout and the accumulated fallout based on a short effective halflife. The values of the coefficients for the two expressions additionally demonstrate the significantly higher transfer of Sr-90 via river runoff compared to that for Cs-137; for the equilibrium situation with a constant fallout rate the river runoff of Sr-90 is about a factor of 20 higher than that of Cs-137. The total runoff for each radionuclide was split between the different Baltic Sea sub-regions according to the sizes of the corresponding catchment areas (based on HELCOM 1993). The catchment areas used are shown in Table 3.1.

The input of Cs-137 from the Chernobyl accident into the Baltic Sea area was estimated to 4.5 PBq (CEC, 1990). Observed data from Denmark, Finland and Russia show that the fallout of Sr-90 from the Chernobyl accident was about 2% relative to that of Cs-137. Table 3.2 shows the estimated inputs of Cs-137 into the various regions.

The reported discharges of Cs-137 and Sr-90 shown in Figs. 3.4 and 3.5 from Sellafield and La Hague have been used (BNFL 1993; CEC 1990). However, only a small fraction of these discharges reach the Baltic Sea. The input to the Baltic Sea has been estimated from model calculations which indicate that about 4% of the discharges from Sellafield are transferred to Kattegat compared to about 8% of the discharges from La Hague. The relative transfer of Cs-137 is lower (about 10% relative difference) than that of Sr-90 because of the chemical differences that result in a higher transfer to sediments of Cs-137 than of Sr-90. Due to the efficient mixing of the upper and lower waters in the Kattegat and the Belt Sea, the main part of the activity of these radionuclides from the two reprocessing plants return to the Skagerrak and only about 1% of the discharges of Cs-137 and Sr-90 are estimated to be transferred to the Baltic Proper.

Data on discharges of Cs-137 and Sr-90 from the nuclear installations bordering the Baltic Sea are shown in the Tables 3.3 and 3.4. These data were collected within HELCOM by the MORS Group (Monitoring of Radioactive Substances in the Baltic Sea), and will be published in a HELCOM report (HELCOM 1995). The data are not complete, particularly not for Sr-90. Table 3.5 presents an overview of the sources considered.



## 4 Model Description

Compartmental or box-model analysis is used to simulate the movement of radionuclides between parts of the marine environment. Box-model analysis assumes instantaneous uniform mixing within each box with rates of transfer across the boundaries of the box being proportional to the inventories of material in the source boxes. The model includes water advection and mixing between adjacent boxes, sedimentation, bioturbation, and bioconcentration.

The box-model analysis uses first order differential equations to describe the transfer of contaminant radionuclides between the boxes. The equations are of the form:

$$\frac{dA_i}{dt} = \sum_{j=1}^n k_{ji} A_j - \sum_{j=1}^n k_{ij} A_i - k_i A_i + Q_i$$

where:  $k_{ii}=0$  for all  $i$ ;  $A_i$  and  $A_j$  are activities (Bq) at time  $t$  in boxes  $i$  and  $j$ ;  $k_{ij}$  and  $k_{ji}$  are rates of transfer ( $y^{-1}$ ) between boxes  $i$  and  $j$ ;  $k_i$  is an effective rate of transfer of activity ( $y^{-1}$ ) from box  $i$  taking into account loss of material from the compartment without transfer to another, for example radioactive decay;  $Q_i$  is a source of input into box  $i$  (Bq); and  $n$  is the number of boxes in the system.

The rates of transfer between the aquatic boxes,  $k_{ij}$  ( $y^{-1}$ ) are related to the volume exchanges,  $R_{ij}$  ( $km^3 y^{-1}$ ) according to:

$$R_{ij} = k_{ij} V_i$$

where  $V_i$  is the volume of water represented by box  $i$ . The present model was modified somewhat from the previous version (Nielsen 1994) by adjusting the mixing conditions (water exchange rates) for some adjacent water boxes of the Baltic Sea while keeping the net advective water fluxes of the model unaltered. The modification was carried out by minimising the difference between the model predictions and observed levels of Cs-137 in the Baltic seawater.

Figure 4.1 shows the regions used in the marine box model near the Baltic Sea, and Figure 4.2 shows a diagrammatic representation of the positions, and interconnections among, individual water boxes for the entire model. Each of the water compartments has associated suspended sediment and the water compartments having seabed boundaries have underlying sediment compartments. The water compartments have odd numbers and the surface sediment compartments have even numbers. The latter are not shown in Figure 4.2.

The compartment names, volumes, mean depths, suspended sediment loads and sedimentation rates are given in Table 4.1. The volume exchange rates used by the model are given in Table 4.2.

At any given time the activity in the water column is partitioned between the water phase and the suspended sediment material. The fraction of the total activity ( $F_w$ ) in the water column that is in aqueous solution is given by:

$$F_w = \frac{1}{1 + K_d SSL}$$

where  $K_d$  is the sediment distribution coefficient ( $m^3 t^{-1}$ ) and SSL the suspended sediment load ( $t m^{-2}$ ). Values for  $K_d$  of 3000 and 1000 are used for Cs-137 and Sr-

90), respectively (IAEA 1985). Activity on suspended sediments is lost to the underlying boxes when particulates settle out. The fractional transfer from a water column (box i) to the sediments (box j) due to sedimentation is given by:

$$k_{ij} = \frac{K_d SR_i}{d_i (1 + K_d SSL_i)}$$

where  $d_i$  (m) is the mean water depth of the water column and  $SR$  ( $t\ m^{-2}\ y^{-1}$ ) the sedimentation rate.

The model includes transfers of radioactivity between the surface sediment layer and the bottom boundary layer. This transfer is represented by diffusivity through the pore water, using a diffusion coefficient of  $1 \cdot 10^{-9}\ m^2\ s^{-1}$ , and mixing due to bioturbation, modelled as a diffusive process using a diffusion coefficient of  $1 \cdot 10^{-12}\ m^2\ s^{-1}$  (NRPB et al. 1994). Removal of activity from the top surface sediment to lower sediment layers is taken into account by assuming that the burial rate is equal to the flux of particles settling from the overlying waters. Radioactive decay is accounted for in all boxes.

## 5 Reliability of Model Predictions

Calculations have been carried out with the model for the time period 1950 to 2100 using the sources specified.

### 5.1 Results for Cs-137

The calculated concentrations of Cs-137 in seawater, sediments and biota are shown in the Figures 5.1.1, 5.1.3 and 5.1.5, respectively. For the water concentrations in Fig. 5.1.1 it is noted that all regions reflect the culmination of the nuclear weapons fallout in the 1960's. In the 1970's and early 1980's the large Sellafield discharges determine the levels in the Kattegat and Belt Sea, but not further into the Baltic Sea. Nearly all regions reflect the impact of the significant Chernobyl fallout in 1986 which is the reason that the Baltic Sea at present in 1994 has the highest Cs-137 concentrations found in seawater anywhere. The observed data are included in the graphs where circles show the annual mean concentrations. It is noted that there generally is a good agreement between the observations and the model predictions. However, in the deep waters of the Eastern Baltic (box 79) the predicted levels differ significantly from the observed values after 1986. The comparison is further illustrated in Fig. 5.1.2 showing a scatterplot between all ( $n=130$ ) observed and predicted concentrations of Cs-137 in seawater. The full line gives the ideal 1:1 relationship, and the points are seen to scatter on both sides of the line. The geometric mean of the predicted-to-observed (P/O) ratio is 1.1 with a geometric standard deviation of 1.4.

The calculated concentrations of Cs-137 in surface sediments are shown in Fig. 5.1.3. The sediment concentrations are seen to respond more slowly to the source terms than the water concentrations. The observed data are much more scarce than for seawater. This reflects the fact that sediment sampling is much more demanding in terms of time and resources compared to water sampling. Furthermore, the variability of marine sediment data is very much larger within a small area than

seawater data due mainly to inhomogeneities in sediments and very different mixing processes in the two media. However, the comparison between observed and predicted values shows a distinct tendency to underpredict the concentrations of Cs-137 in surface sediments after the Chernobyl accident. Figure 5.1.4 shows a scatterplot of the observed and predicted sediment concentrations ( $n=35$ ). The geometric mean of the P/O ratio is 0.3 with a geometric standard deviation of 2.4.

The calculated concentrations of Cs-137 in biota are shown in Fig. 5.1.5. For Kattegat (box 71) calculations are done for both molluscs and fish, and for the remaining regions for fish only. The biota concentrations are calculated from concentrations of Cs-137 in filtered seawater using recommended bioconcentration factors ( $200 \text{ Bq l}^{-1}$  fish per  $\text{Bq m}^{-3}$  water, and  $30 \text{ Bq l}^{-1}$  molluscs per  $\text{Bq m}^{-3}$  water; Koivulehto and Saxén, 1981; IAEA, 1985). Figure 5.1.6 shows a scatterplot of the observed and the predicted values ( $n=89$ ). The geometric mean of the P/O-ratios is 1.7 with a geometric standard deviation of 2.3.

The inventory of Cs-137 in the Baltic Sea has been calculated with the model as a function of time. Figure 5.1.7 shows the inventory broken down according to the two compartments: water and sediments. The transfer of Cs-137 from water to the sediments with time is illustrated. The results show that the inventory is approximately equally divided between water and sediments before the Chernobyl accident in 1986 after which most of the activity is found in the water. These predicted data are shown in Fig. 5.1.8 together with observed data from Salo et al. (1985).

It is noted that there is a good agreement for the water data prior to 1986 after which the model overestimates the water levels. This could indicate a different (faster) rate of transfer from the water to the sediments of the Cs-137 from the Chernobyl fallout compared to that from the atmospheric fallout from the nuclear weapons testing. This is in qualitative agreement with the comparison between the observed and predicted sediment concentrations after the Chernobyl accident. For all the water inventory data ( $n=32$ ) the geometric mean P/O ratio is 1.2 with a geometric standard deviation of 1.2. For the sediment data there is a reasonable agreement between observations and predictions, but the number of observations are few ( $n=6$ ). The geometric mean of the P/O ratios for the sediment inventory data is 0.9 with a geometric standard deviation of 1.7.

Figure 5.1.9 shows the Cs-137 inventory broken down according to the four sources: nuclear weapons fallout, Chernobyl fallout, discharges from reprocessing facilities and discharges from nuclear installations at the Baltic Sea. The graph is shown with a logarithmic vertical scale in order to cover the range from the total inventory to that from the nuclear installations which is more than three orders of magnitude lower than the former. The two major sources to the inventory are the nuclear weapons fallout and the Chernobyl fallout in agreement with the inputs shown in Table 3.5. The Cs-137 inventory is estimated in 1994 at a value of about 3200 TBq of which 11% originate from nuclear weapons fallout, 84% from the Chernobyl accident, 5% from European reprocessing and 0.04% from nuclear installations located in the Baltic Sea area.

## 5.2 Results for Sr-90

Calculations have been carried out for Sr-90 as for Cs-137. The calculated water concentrations of Sr-90 are shown in Fig. 5.2.1 together with the observed values. The same tendencies are seen as for Cs-137: the fallout from the nuclear weapons testing is dominating the levels that peak in the early 1960's, the influence of discharges from the European reprocessing plants are only discernible in the Kattegat and the Belt Sea, and the contribution from the Chernobyl accident in

1986 is apparent but relatively much lower than for Cs 137. The observed data generally compare well with the predicted values. It is noted that the predicted values for the deep waters of the Eastern Baltic (box 79) are low compared to the observed values in agreement with the comparison for the Cs-137 data for that box. This indicates a need for improving the model by adjusting the water exchanges between that and the adjacent water regions. The influence of the Chernobyl fallout on the concentrations of Sr-90 in the water is obvious from the predicted values but not equally so from the observed values. However, the comparison between observed and predicted values may be interpreted as a support of the assumptions concerning the input of Chernobyl Sr-90 fallout into the Baltic Sea. Fig. 5.2.2 shows a scatterplot of the observed and predicted levels of Sr-90 in seawater. The geometric mean of the P/O values ( $n=67$ ) is 0.8 with a geometric standard deviation of 1.4.

The calculated concentrations of Sr-90 in surface sediments in the various regions of the Baltic are shown in Fig. 5.2.3. The observed values are few ( $n=10$ ) but indicate a tendency by the model to overpredict the concentrations. The geometric mean P/O ratio is 2.6 with a standard deviation of 2.5.

The calculated concentrations of Sr-90 in fish are shown in Fig. 5.2.4. The calculated concentrations are based on a bioconcentration factor of 2 Bq l<sup>-1</sup> fish per Bq m<sup>-3</sup> filtered seawater (IAEA 1985). It is noted that the predicted levels fall in two groups that either significantly overestimates or underestimates the observed levels. This is also apparent in Fig. 5.2.5 showing a scatterplot between the observed and predicted values. This indicates that the single bioconcentration factor used for the calculations in this case may be too simplistic a concept. However, the model may be acceptable for the average conditions since the geometric mean of the P/O ratios is 0.9 which is close to unity; but the geometric standard deviation is large, 4.4.

The inventory of Sr-90 in the Baltic Sea is shown in Fig. 5.2.6 which gives the inventories in water and sediment as a function of time. It is noted that the relative proportion of Sr-90 in the sediments is estimated to be smaller than for Cs-137. Fig. 5.2.7 shows a comparison between the observed (data from Salo et al., 1985) and predicted inventory of Sr-90 in the Baltic seawater. The agreement is good and the geometric mean of the P/O ratios is 1.0 with a geometric standard deviation of 1.2. The total inventory is shown in Fig. 5.2.8 broken down according to the four sources: weapons fallout, Chernobyl fallout, reprocessing and nuclear installations. The dominating source is fallout from nuclear weapons testing. The inventory is estimated at a level of 370 TBq Sr-90 in 1994 of which 75% originate from weapons fallout, 18% from the Chernobyl accident, 7% from European reprocessing facilities, and 0.003% from nuclear facilities located in the Baltic Sea area.

## 6 Collective Doses

### 6.1 Assumptions Concerning Seafood Consumption

Total catches of fish, crustacea and molluscs were taken into account and assumed to be used for human consumption. The fisheries statistics from 1991 were obtained from the International Council for the Explorations of the Seas (ICES 1993) and are shown in Table 2.1. The fish catch is dominated by the species herring (60%), cod (20%), sprat (16%), and the remainder concerns flounder, salmon, dab, trout, turbot and plaice. For the calculations no distinction has been

made between fish species. The total landings shown in Table 2.1 have been used for the calculations combined with relative fractions of the total landings assumed to be actually eaten by man. For these fractions the following values were used: one half for fish, one third for crustacea and one sixth for molluscs. It has furthermore been assumed that the annual harvest of marine produce has remained unaltered equal to that shown in Table 2.1 throughout the time period considered.

## 6.2 Collective Dose Rates and Collective Doses

The present study does not include an identification or grouping of the human populations exposed to radioactivity from ingestion of seafood from the Baltic Sea area, and assumes that all marine produce is consumed by man.

The results for the collective dose rates are shown in Figs. 6.2.1 and 6.2.2 giving the collective dose rates broken down in components according to sources and nuclides. The collective dose rate is estimated to reach a maximum of about 42 manSv  $y^{-1}$  in the 1960's before 1986 where the collective dose rate peaks at about 160 manSv  $y^{-1}$  due to the Chernobyl accident. The time course of the dose rate from weapons fallout is predicted to be reduced at a rate slower than that from the other sources after year 2000 due to inflow of radionuclides to the Baltic Sea from the North Atlantic via the North Sea.

The collective committed doses are given in Table 6.2.1. The collective committed dose from Cs-137 and Sr-90 is thus estimated at 2300 manSv of which about 1% is estimated to be due to Sr-90 and the rest to Cs-137. The main contribution of the dose from Sr-90 is due to fallout from nuclear weapons where transfer from the catchment area to the sea via river runoff gives a significant contribution.

The dominating source to the collective committed dose is the fallout from the Chernobyl accident which is estimated to contribute 1400 manSv corresponding to about 60% of the total. Then follows the atmospheric fallout from nuclear weapons testing estimated to give rise to a collective dose of about 700 manSv corresponding to about 30% of the total. Discharges from European nuclear reprocessing plants are estimated to cause a committed collective dose of 200 manSv in the Baltic Sea area corresponding to about 10% of the total, and discharges from nuclear installations bordering the Baltic Sea area are estimated to cause 0.6 manSv corresponding to about 0.03% of the total.

In the Marina project (CEC 1990) estimates were made of collective doses from ingestion of Cs-137 in seafood from Chernobyl fallout into the Baltic Sea. Two methods were applied, one based on mean residence times of conservative substances and radiocaesium inventories in the seawater and another using a box model similar to the present one, but without considering sedimentation. The committed doses were estimated at 2400 manSv and 3800 manSv by the two methods, respectively. These values are significantly higher than the present estimate of 1400 manSv. But if one considers the differences between the assumptions made (source term, mean residence time and fishery statistics), the first method agrees well (within 15%) with the present estimate. The second method still gives an estimate well (about a factor of two) above the present one, but this difference is explained by lack of sedimentation in that box model.

Furthermore, it should be noticed that the present collective dose estimate from the Chernobyl accident is probably on the high side. The dose depends mainly on the time course from 1986 and onwards of the concentration of Cs-137 in the Baltic seawater. From the comparison made in Section 5.1 of the measured and calculated inventories of Cs-137 in the Baltic seawater it is noted, that the model underestimates the rate of reduction after the Chernobyl accident in 1986. The rate

of reduction of the model corresponds to a mean residence time of 8 years whereas the rate of reduction from the observed data corresponds to a mean residence time of 3 years ( $\pm 25\%$ , 1 sd). However, the observed data cover five years only, and a longer time of observation is needed for a more reliable estimate of the rate of reduction which additionally may not be adequately described by a single exponential function (a mean residence time) but rather by two exponential terms. So in order to improve the estimate of the collective doses from seafood ingestion of Cs-137 from the Chernobyl accident it is important that the observed data for Cs-137 in Baltic seawater are updated and followed for a longer time period for a more reliable estimate of the rate of reduction.

### 6.3 Comparison with Doses from Natural Radionuclides

Naturally-occurring radionuclides in seawater are incorporated into marine organisms and give rise to radiation exposure to humans from ingestion of seafood. Polonium-210 is the major contributor in this context due to the high bio-accumulation in marine organisms and the relatively high dose factor. Typical Po-210 concentrations in fish from the Baltic Sea are  $0.8 \text{ Bq kg}^{-1}$  (Bojanowsky 1994; Dahlgaard 1994; Holm 1994), in crustacea  $20 \text{ Bq kg}^{-1}$  (Swift et al. 1994) and in molluscs  $30 \text{ Bq kg}^{-1}$  (Dahlgaard 1994). From the same assumptions of seafood intake as in the preceding sections and using a dose factor of  $6.2 \cdot 10^{-7} \text{ Sv Bq}^{-1}$  (Phipps et al. 1991) the collective dose rate from Po-210 may be calculated using the data on fishery statistics in Table 2.1 resulting in a value of about  $200 \text{ manSv y}^{-1}$ . This is slightly higher than the peak value of the collective dose rate from Cs-137 and Sr-90 in 1986 ( $160 \text{ manSv y}^{-1}$ ).

## 7 Conclusions

A model has been developed that enables assessments of the radiological consequences of releases of radioactivity into the Baltic Sea to be carried out. The model includes the dispersion of radionuclides in the marine environment, the transfer of radionuclides to biota, and the calculation of doses to individuals and populations exposed to radionuclides in seafood.

The dispersion model is based on box-model analysis and includes 12 water and 12 sediment boxes for the Baltic Sea area. The physical processes covered by the model are net advection and mixing of water between adjacent boxes, sedimentation of particulate material from the water column to the top sediment, and biological mixing in the top sediment. The model is intended for the prediction of annual average concentrations of radionuclides in the marine environment.

Concentrations of radionuclides are calculated by the model from radionuclide concentrations in filtered seawater using recommended concentration factors.

The quality of the model predictions has been investigated by comparing predicted levels of Cs-137 and Sr-90 in water, top sediment and biota with observed levels. The observed data originate partly from the HELCOM database (HELCOM 1995) and partly from published material and cover the time period from 1960 to 1992. The sources of radioactivity into the Baltic region that have been considered are fallout from atmospheric nuclear weapons testing, fallout from the Chernobyl accident in 1986, discharges of radionuclides from the two European reprocessing

plants (Sellafield and La Hague) transported into the Baltic Sea, and discharges of radionuclides from nuclear installations located in the Baltic Sea area.

The comparisons have been made in terms of predicted-to-observed (P/O) ratios for which geometric means and geometric standard deviations have been calculated. The geometric mean of the P/O-ratio is an overall factor that indicates the predictive accuracy by how much the model overestimates the observed data (P/O-ratio higher than unity) or underestimates the observed data (P/O-ratio lower than unity). Similarly, the geometric standard deviation indicates the predictive precision by a single factor within which 68% of the values vary around the geometric mean value. From the geometric standard deviation another factor may be calculated that corresponds to the variability of 95% of the values. In the following this factor is used for the predictive precision.

The predicted water concentrations are generally in good agreement with the observations for both radionuclides; the factor of predictive accuracy is close to unity and the factor of predictive precision is below 2. For the top sediment concentrations the agreement is less favourable; for Cs-137 the model underestimates the observed levels with about a factor of 3 and for Sr-90 the model overestimates with about a factor of 3, and the factor of predictive precision is about 6 for both isotopes. For the biota data the comparison shows that the model overestimates the Cs-137 levels with about a factor of 1.7 combined with a factor of predictive precision of about 5, whereas the predictive accuracy for Sr-90 is fine but with a very large factor of predictive precision of 20. The predicted water inventory data compare well with the observed data for the two radionuclides both in terms of predictive accuracy and predictive precision. For the Cs-137 sediment inventory data the predictive precision is also good, but not as good as for the water data.

The inventories of Cs-137 and Sr-90 in the Baltic Sea in 1994 have been estimated at 3200 TBq and 370 TBq, respectively. For both radionuclides most of the activity is in the water while the rest is in the bottom sediments. For Cs-137 the inventory is due mainly to fallout from the Chernobyl accident (84%), while the rest originates from nuclear weapons fallout (11%), discharges from European reprocessing plants (5%) and discharges from nuclear facilities located in the Baltic Sea area (0.04%). For Sr-90 the main source of contamination is fallout from nuclear weapons testing (75%), while the rest is due to fallout from the Chernobyl accident (18%), discharges from the European reprocessing plants (7%) and discharges from nuclear facilities located in the Baltic Sea area (0.003%).

Collective dose rates and doses to members of the public have been calculated based on fishery statistics and on predicted concentrations of Cs-137 and Sr-90 in biota. The collective dose rate peaks in 1986 at a level of 160 manSv y<sup>-1</sup> due to fallout from the Chernobyl accident. The total collective dose from Cs-137 and Sr-90 in the Baltic Sea is estimated at 2300 manSv of which about 60% originates from Chernobyl fallout, about 30% from fallout from nuclear weapons testing, about 10% from European reprocessing facilities, and about 0.03% from nuclear installations bordering the Baltic Sea area.

Doses from naturally-occurring radioactivity in seafood (polonium-210) have been calculated on a similar basis and compared with the doses from Cs-137 and Sr-90. The results of this comparison are shown in the Tables 7.1 and 7.2 which summarise the calculated dose rates and doses. It is noted that dose rates and doses from natural radioactivity dominate except for the year 1986 where the collective dose rate from Chernobyl fallout approached that from natural radioactivity.

The reliability of the model calculations of Cs-137 in biota, which are important for the dose calculations, was tested and showed a predictive accuracy slightly less than 2. No formal uncertainty analysis has been carried out, but it is estimated that the predictive accuracies of the calculated dose rates and doses are about

a factor of 3.

The present work represents a first step towards a full assessment of the exposure of members of the public to radionuclides in the Baltic Sea. A full assessment should include a more comprehensive list of radionuclides and exposure pathways and more complete data on radioactive discharges to sea from nuclear facilities bordering the Baltic Sea.



## TABLES

*Table 2.1. Catches of fish, crustacea and molluscs (metric tonnes) in the Baltic Sea areas in 1991 (ICES 1993).*

Area	Fish (t)	Crustacea (t)	Molluscs (t)
Kattegat	66000	988	375
Belt Sea	40989	143	11034
Baltic Sea West	215151		
Baltic Sea East	232638		
Bothnian Sea	30797		
Bothnian Bay	7739		
Gulf of Finland	39941		
Total	633255	1131	11409

*Table 3.1. Catchment areas for the sub-regions of the Baltic Sea used to calculate runoff.*

Baltic Sea Regions	Catchment areas	
	(km <sup>2</sup> )	(%)
Kattegat	87400	5.1
Belt Sea	4300	0.3
Baltic Sea West	108250	6.3
Baltic Sea East	469600	27.3
Bothnian Sea	228100	13.3
Bothnian Bay	277000	16.1
Gulf of Finland	413300	24.0
Gulf of Riga	132100	7.7
Total	1720050	100

*Table 3.2. Direct fallout Cs-137 and Sr-90 to Baltic Sea water regions from the Chernobyl accident.*

Baltic Sea Regions	Cs-137 (TBq)	Sr-90 (TBq)
Belt Sea	60	1.2
Baltic Sea West	980	19.6
Baltic Sea East	490	9.8
Bothnian Sea	2400	48
Bothnian Bay	210	4.2
Gulf of Finland	320	6.4
Gulf of Riga	40	0.8
Total	4500	90

*Table 3.3. Discharges to sea of Cs-137 (GBq/y, rounded) from nuclear installations bordering the Baltic Sea.*

Year	Kattegat	Belt Sea	Baltic Sea West	Baltic Sea East	Bothnian Sea	Gulf of Finland	Gulf of Riga	Total
1984	9.2	6.2	160	1.1	0.4	9.3		180
1985	1.8	3.4	89	1.2	0.4	70		170
1986	5.0	15	43	1.8	0.5	100		170
1987	0.09	7.7	35	0.7	0.6	46		90
1988	0.6	12	31	1.6	0.4	69		110
1989	1.0	8.2	22	0.2	0.2	530		560
1990	3.2	14	14	0.5	0.1	330	0.02	360
1991	6.7	19	25	1.9	0.8	15	0.01	68
Total	28	86	420	9.0	3.5	1200	0.03	1700

*Table 3.4. Discharges to sea of Sr-90 (GBqly, rounded) from nuclear installations bordering the Baltic Sea.*

Year	Kattegat	Belt Sea	Baltic Sea West	Baltic Sea East	Bothnian Sea	Gulf of Finland	Gulf of Riga	Total
1984				0.04	0.003	0.001		0.05
1985				0.02				0.015
1986						0.002		0.002
1987								
1988						0.002		0.002
1989		0.01	8.2	0.01	0.01	0.16		8.4
1990				5.9		0.001	0.005	5.9
1991				0.003			0.009	0.012
Total		0.01	8.2	6.0	0.02	0.17	0.014	14

*Table 3.5. Discharges of Cs-137 and Sr-90 to the Baltic Sea in 1950 to 1994.*

Source	Cs-137 (TBq)	Sr-90 (TBq)
Weapons fallout, direct deposition to sea	1780	1130
Weapons fallout, river runoff	70	410
Chernobyl fallout	4500	90
European reprocessing	380	70
Nuclear installations	1.7	0.014
Total	6732	1700

*Table 4.1. Compartment names, volumes, mean depths, suspended sediment loads and sedimentation rates.*

Box	Name	Volume (m <sup>3</sup> )	Depth (m)	SSL (t m <sup>-1</sup> )	SR (t m <sup>2</sup> y <sup>-1</sup> )
1	Other Oceans	1.0E+18	4.0E+03	1.0E-07	5.2E-05
3	Atlantic Ocean	3.0E+17	3.5E+03	1.0E-07	1.0E-04
5	North-East Atlantic	5.0E+16	3.5E+03	1.0E-07	1.0E-05
7	Arctic Ocean	1.7E+16	1.2E+03	1.0E-07	1.0E-05
9	Spitsbergen Waters	1.0E+14	1.2E+03	1.0E-07	1.0E-05
11	Barents Sea	3.0E+14	2.0E+02	1.0E-07	1.0E-05
13	Norwegian Coastal Waters	1.0E+15	1.2E+03	1.0E-07	1.0E-05
15	Scottish Waters West	1.0E+13	1.1E+02	1.0E-06	1.0E-04
17	Scottish Waters East	3.0E+12	1.1E+02	1.0E-06	1.0E-04
19	Irish Sea North West	4.1E+11	9.3E+01	3.0E-06	3.0E-03
21	Irish Sea North	6.0E+10	3.4E+01	3.0E-06	5.1E-03
23	Irish Sea North East	5.2E+10	2.4E+01	3.0E-06	3.6E-03
25	Irish Sea West	6.6E+11	6.3E+01	3.0E-06	2.0E-03
27	Irish Sea South East	1.6E+11	3.1E+01	3.0E-06	4.7E-03
29	Cumbrian Waters	3.8E+10	2.8E+01	3.0E-06	4.2E-03
31	Irish Sea South	1.1E+12	5.7E+01	1.0E-06	1.0E-04
33	Liverpool and Morecambe Bays	3.2E+10	1.3E+01	3.0E-06	2.0E-03
35	Celtic Sea	2.0E+13	1.5E+02	1.0E-06	1.0E-04
37	Bristol Channel	1.0E+12	5.0E+01	1.0E-06	1.0E-04
39	Bay of Biscay	6.5E+14	4.0E+03	1.0E-06	1.0E-05
41	French Continental Shelf	3.5E+13	3.5E+02	5.0E-07	1.0E-04
43	Cantabrian Sea	3.0E+13	7.6E+02	1.0E-06	2.0E-04
45	Portuguese Continental Shelf	1.5E+13	4.9E+02	1.0E-06	2.0E-04

Table 4.1. Continued.

Box	Name	Volume (m <sup>3</sup> )	Depth (m)	SSL (t m <sup>-1</sup> )	SR (t m <sup>-2</sup> y <sup>-1</sup> )
47	Gulf of Cadiz	2.3E+14	1.7E+03	2.0E-07	5.0E-05
49	Mediterranean Sea	4.0E+15	1.3E+03	1.0E-06	1.0E-04
51	English Channel West	3.2E+12	6.0E+01	1.0E-06	1.0E-04
53	English Channel South East	6.5E+11	4.0E+01	1.0E-06	1.0E-04
55	English Channel North East	6.5E+11	4.0E+01	1.0E-06	1.0E-04
57	North Sea South West	4.5E+11	3.1E+01	6.0E-06	1.6E-04
59	North Sea South East	9.5E+11	3.7E+01	6.0E-06	1.9E-04
61	North Sea Central	1.3E+13	5.0E+01	6.0E-06	1.0E-04
63	North Sea East	1.2E+12	2.2E+01	6.0E-06	4.4E-05
65	North Sea North	5.6E+13	2.4E+02	6.0E-06	1.0E-04
67	Skagerrak	6.8E+12	2.1E+02	1.0E-06	7.5E-04
69	Kattegat, deep	2.0E+11	1.0E+02	1.0E-06	7.5E-04
71	Kattegat, surface	3.2E+11	2.0E+01	1.0E-06	7.5E-04
73	Belt Sea, deep	1.4E+11	3.0E+01	1.0E-06	7.5E-04
75	Belt Sea, surface	1.5E+11	1.4E+01	1.0E-06	7.5E-04
77	Baltic Sea West, deep	7.7E+11	1.1E+02	3.0E-07	7.5E-04
79	Baltic Sea East, deep	1.5E+12	1.1E+02	3.0E-07	7.5E-04
81	Baltic Sea West, surface	3.8E+12	4.9E+01	3.0E-07	7.5E-04
83	Baltic Sea East, surface	7.0E+12	5.3E+01	3.0E-07	7.5E-04
85	Bothnian Sea	4.9E+12	6.2E+01	3.0E-07	1.5E-03
87	Bothnian Bay	1.5E+12	4.1E+01	3.0E-07	1.0E-03
89	Gulf of Finland	1.1E+12	3.7E+01	3.0E-07	1.5E-03
91	Gulf of Riga	4.1E+11	2.3E+01	3.0E-07	7.5E-04

Table 4.2. Exchange rates between boxes ( $\text{km}^2 \text{ y}^{-1}$ ).

From box	To box	Exchange rate ( $\text{km}^2 \text{ y}^{-1}$ )	From box	To box	Exchange rate ( $\text{km}^2 \text{ y}^{-1}$ )	From box	To box	Exchange rate ( $\text{km}^2 \text{ y}^{-1}$ )
1	3	1000000	17	15	500	31	27	75
3	1	1000000	17	61	8000	31	35	600
3	5	500000	17	65	2400	33	27	109
5	3	500000	19	15	7400	33	29	55
5	9	100000	19	21	333	35	5	4600
5	11	100000	19	25	514	35	31	3000
5	13	14500	21	19	833	35	37	2000
5	15	8300	21	23	183	35	39	150000
5	35	10000	21	27	173	35	41	140000
5	39	670000	23	21	238	35	51	7000
5	43	109000	23	29	100	37	35	2000
5	45	460000	25	19	2414	39	5	670000
5	47	510000	25	27	933	39	35	150000
5	65	34000	25	31	600	39	41	580000
7	5	240000	27	21	568	39	43	390000
9	5	20000	27	25	433	41	35	140000
9	7	220000	27	29	230	41	39	580000
11	7	20000	27	31	75	41	43	74000
11	9	140000	27	33	129	43	5	109000
13	11	60000	29	23	205	43	39	390000
15	5	500	29	27	145	43	41	75000
15	17	10700	29	33	35	43	45	15000
15	19	5000	31	25	3000	45	5	460000

Table 4.2. continued.

From box	To box	Exchange rate (km <sup>3</sup> y <sup>-1</sup> )	From box	To box	Exchange rate (km <sup>3</sup> y <sup>-1</sup> )	From box	To box	Exchange rate (km <sup>3</sup> y <sup>-1</sup> )
45	43	13000	61	57	696	73	83	270
45	47	60000	61	59	0	75	71	1170
47	5	510000	61	63	9735	75	73	700
47	45	58000	61	65	6908	77	75	220
47	49	52890	61	67	5600	77	79	220
49	47	50580	63	59	2417	77	81	198
51	35	2000	63	61	8700	79	77	440
51	53	3500	63	67	5892	79	83	208
51	55	3500	65	5	1730	81	77	198
53	51	1000	65	13	45400	81	75	720
53	55	300	65	17	0	81	83	4124
53	59	6000	65	61	0	83	79	208
55	51	1000	65	67	4983	83	81	4118
55	53	1300	67	61	5500	83	85	525
55	59	50	67	63	0	83	89	8
57	59	609	67	65	11505	83	91	312
57	61	381	67	69	1545	85	81	970
59	53	50	69	71	930	85	87	372
59	55	1000	69	73	720	87	85	472
59	57	294	71	67	2020	89	83	133
59	61	458	71	69	100	91	83	344
59	63	7274	73	75	930			
61	17	0	73	79	220			

**Table 6.2.1. Collective committed doses to members of the public from ingestion of Cs-137 and Sr-90 in seafood from the Baltic Sea area calculated up to the year 2100.**

Source	Cs-137 (manSv)	Sr-90 (manSv)	Total (manSv)
Chernobyl	1387	1	1388
Weapons fallout	650	18	668
Reprocessing plants	194	1	195
Nuclear installations	0.6	0.0002	0.6
Total	2232	20	2252

**Table 7.1. Summary of maximum dose rates from Cs-137, Sr-90 and Po-210 to the population around the Baltic Sea.**

Sources	Collective dose rate peak (year) (manSv y <sup>-1</sup> )
Chernobyl fallout	160 (1986)
Nuclear weapons fallout	42 (1966)
European reprocessing plants	12 (1982)
Baltic nuclear installations	0.06 (1991)
Natural radioactivity (Po-210)	200

**Table 7.2. Collective exposure of the population around the Baltic Sea for the period from 1950 to 2050 from Cs-137, Sr-90 and Po-210 in Baltic seawater.**

Sources	Collective dose to year 2050 (manSv)
Chernobyl fallout	1400
Nuclear weapons fallout	700
European reprocessing plants	200
Baltic nuclear installations	0.6
Natural radioactivity (Po-210)	20 000



# NUCLEAR WEAPONS FALLOUT

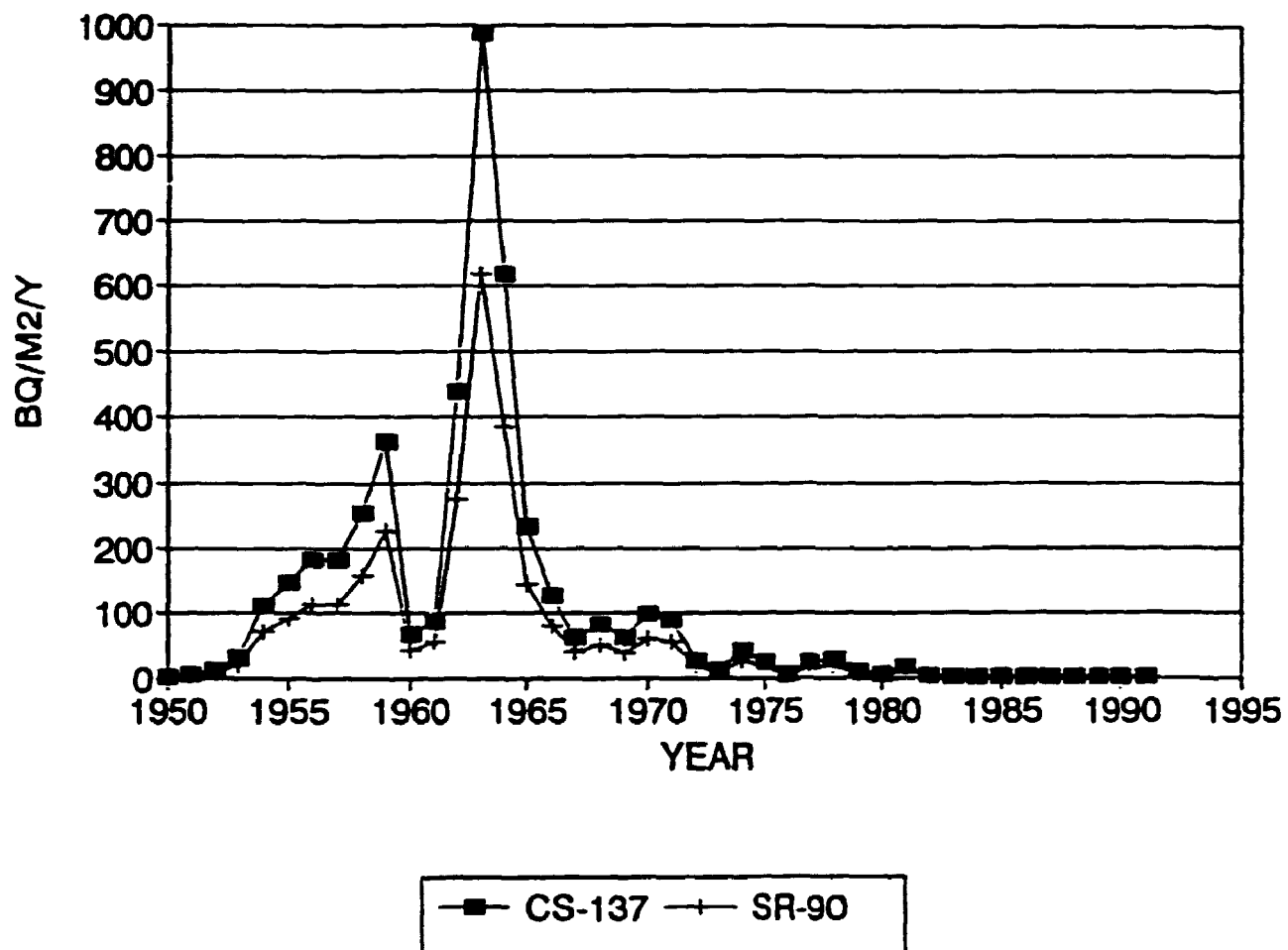
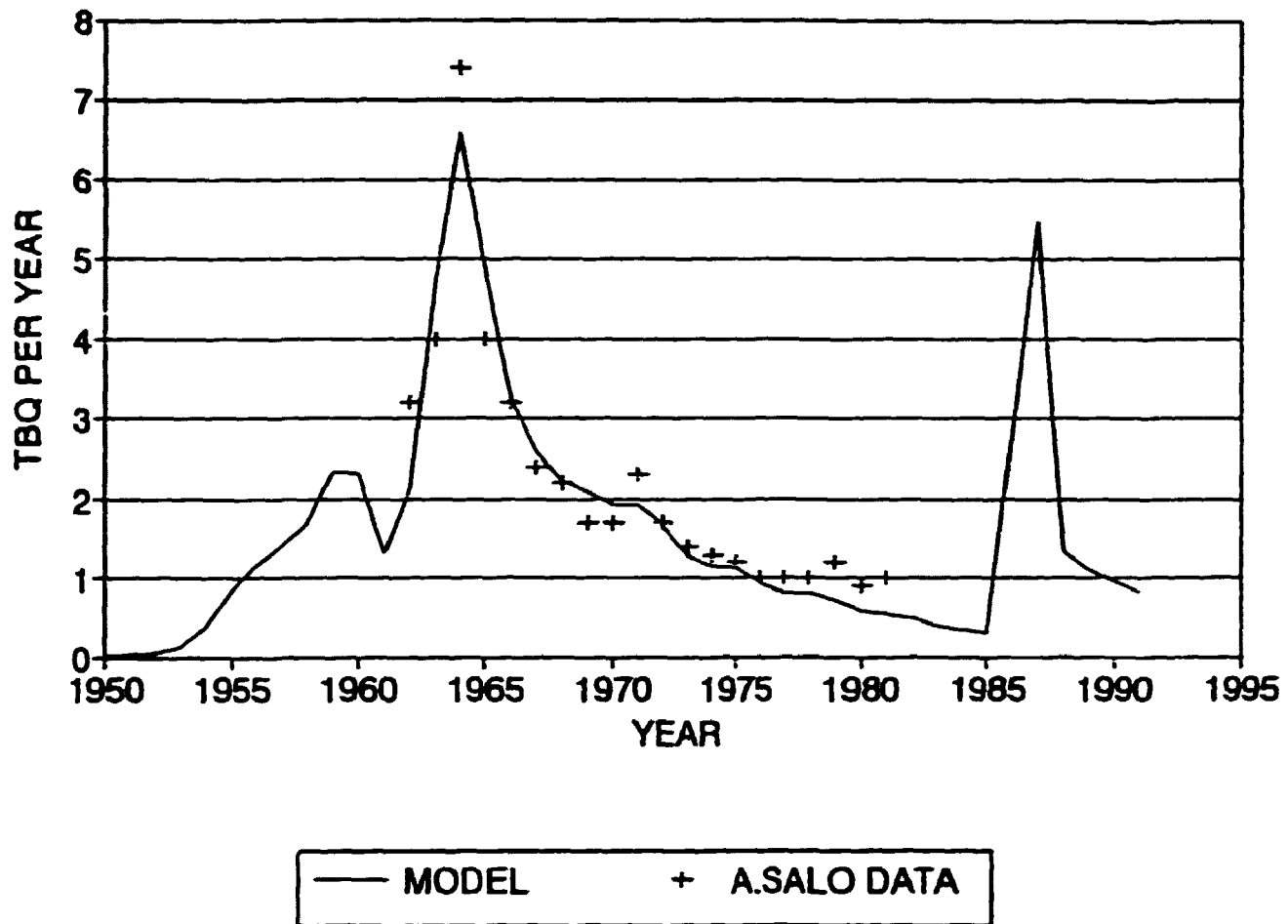


Figure 3.1. Atmospheric fallout of Cs-137 and Sr-90 in Denmark ( $Bq m^{-2}$ ) from nuclear weapon tests.

# CS-137 RIVER RUNOFF TO BALTIC SEA



*Figure 3.2. Predicted and observed runoff (TBq y<sup>-1</sup>) of Cs-137 via rivers to the Baltic Sea shown as a function of time.*

# SR-90 RIVER RUNOFF TO BALTIC SEA

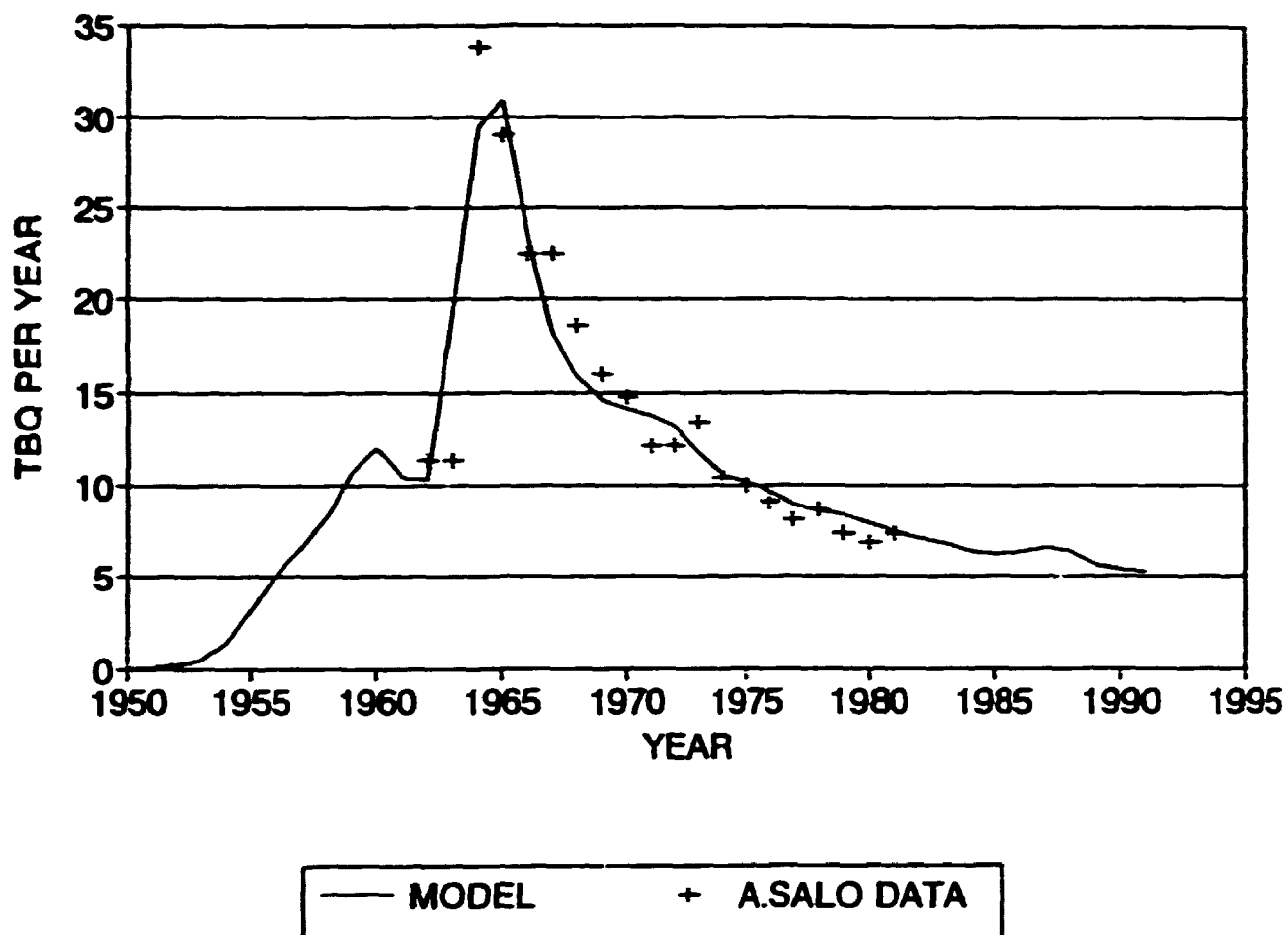


Figure 3.3. Predicted and observed runoff (TBq y<sup>-1</sup>) of Sr-90 via rivers to the Baltic Sea shown as a function of time.

# CS-137 DISCHARGES TO SEA FROM REPROCESSING

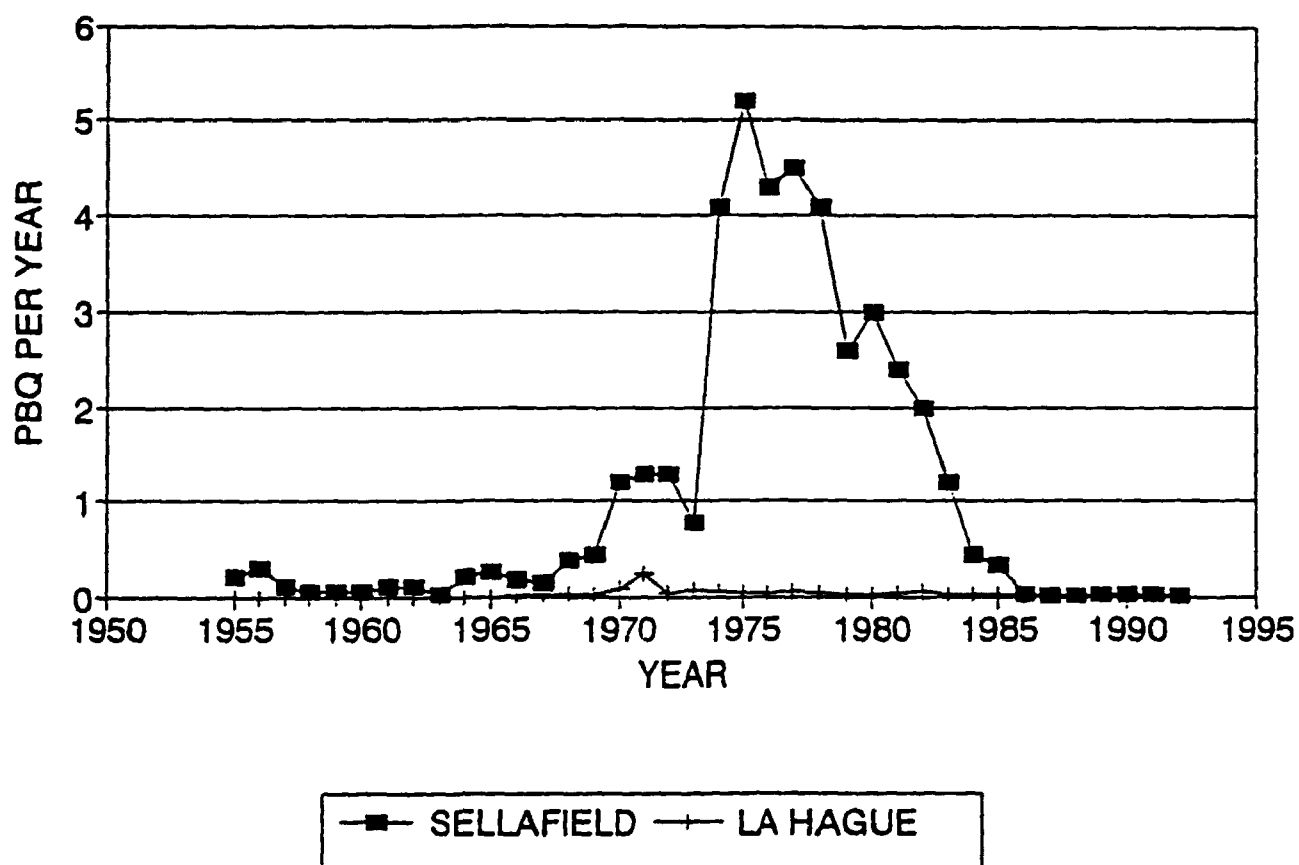


Figure 3.4. Discharges of Cs-137 to sea (PBq y<sup>-1</sup>) from the nuclear reprocessing plants Sellafield in the UK and La Hague in France.

# SR-90 DISCHARGES TO SEA FROM REPROCESSING

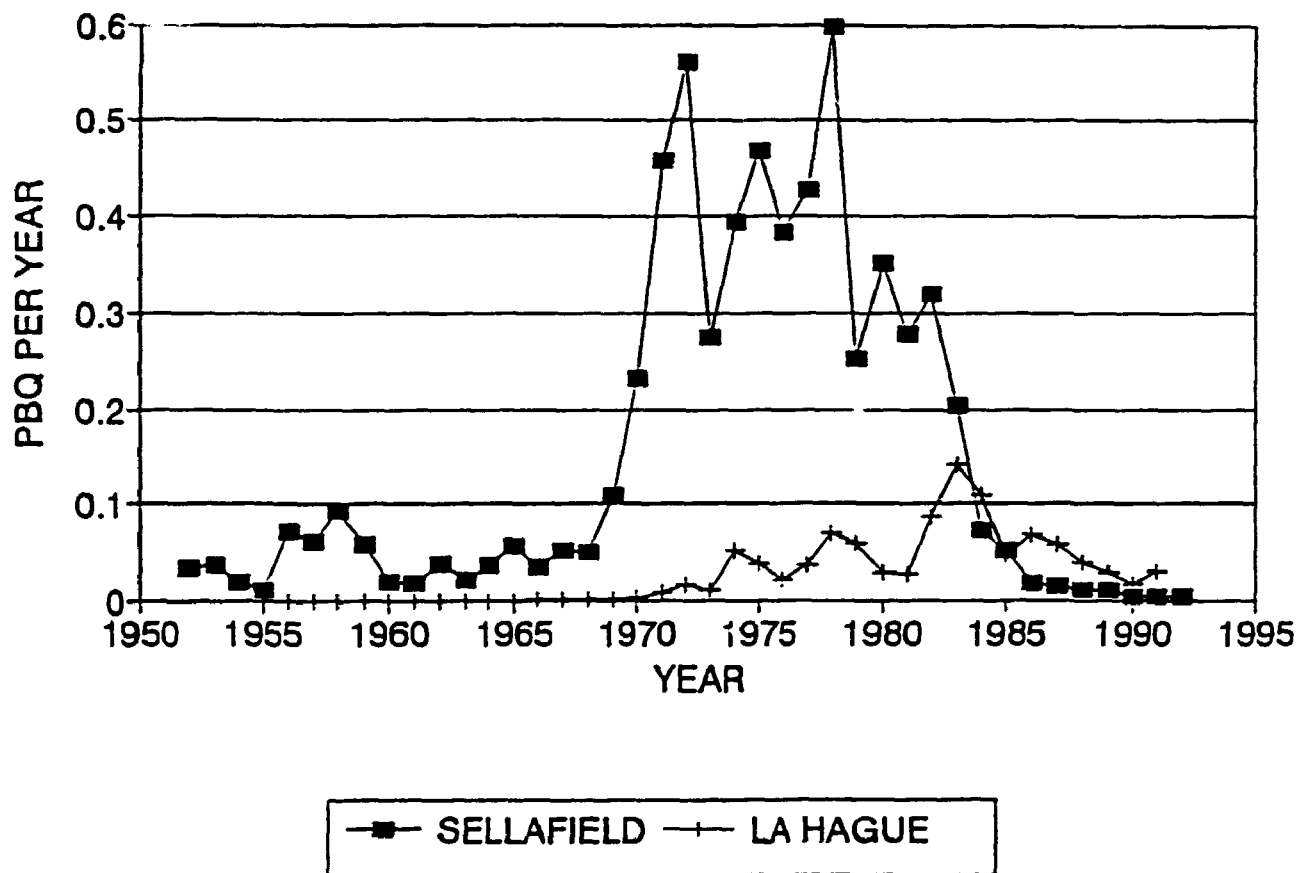
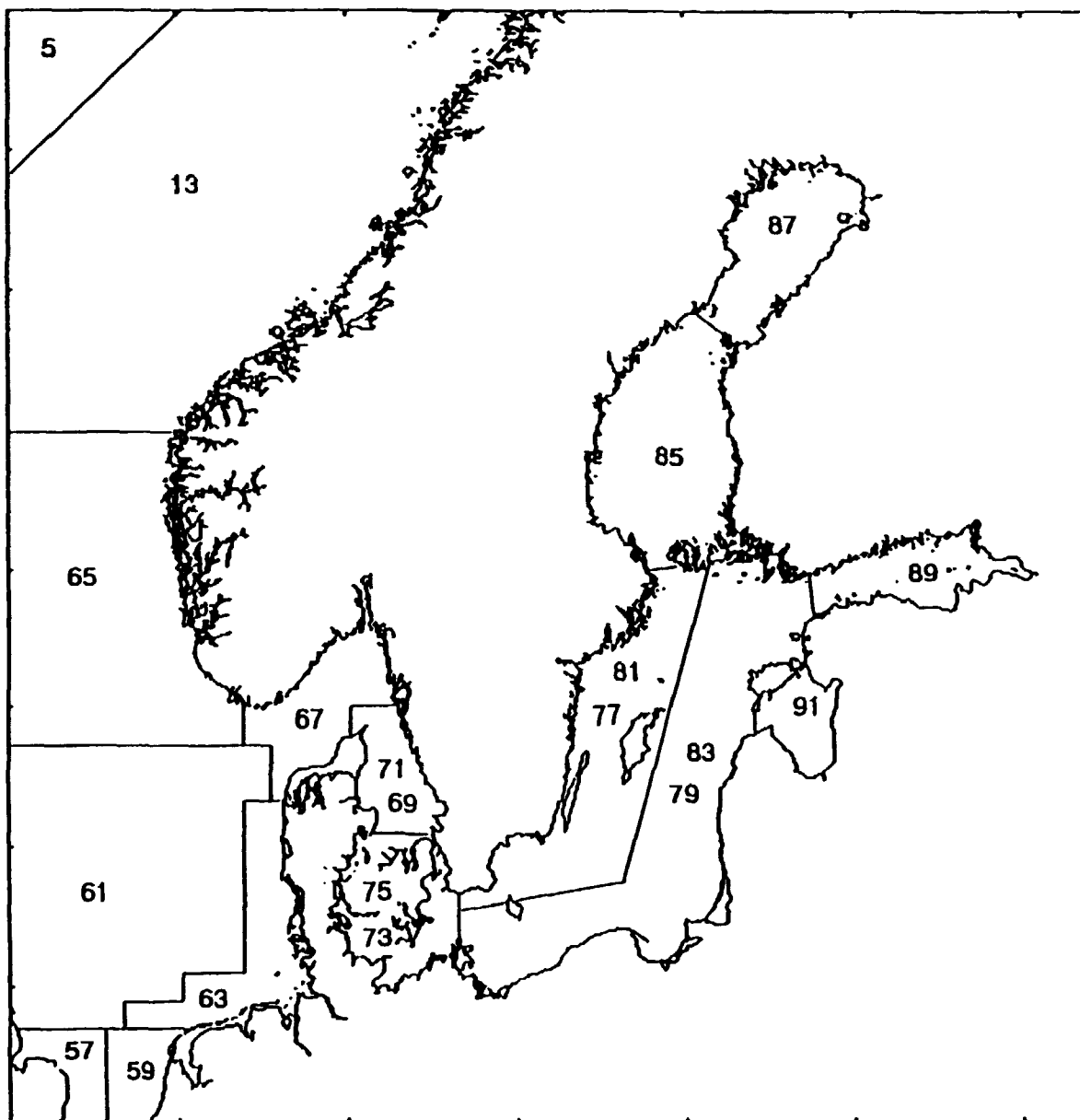


Figure 3.5. Discharges of Sr-90 to sea (PBq y<sup>-1</sup>) from the nuclear reprocessing plants Sellafield in the UK and La Hague in France.



*Figure 4.1. Regions around the Baltic Sea covered by the marine box model. The water-box numbers are indicated.*

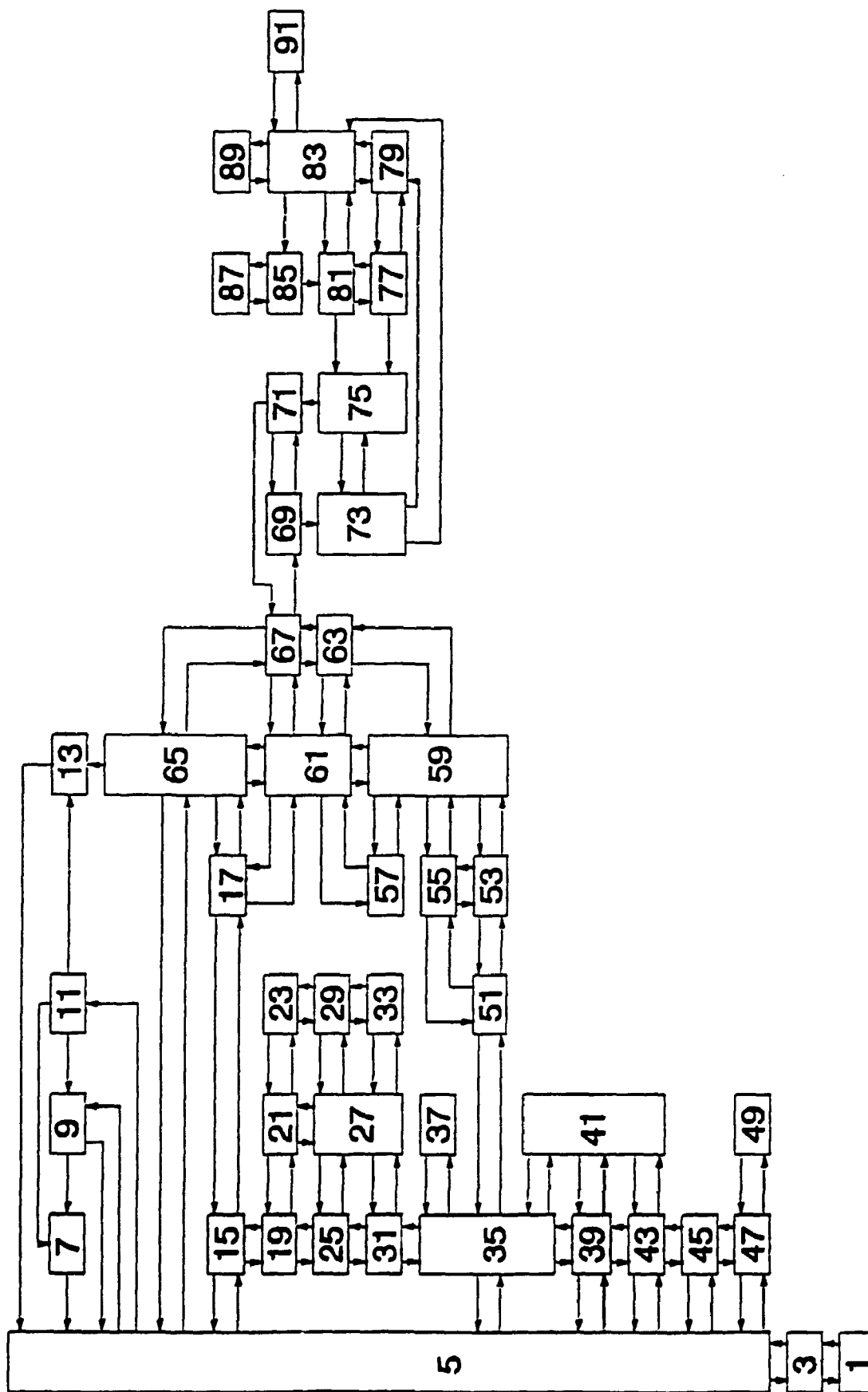


Figure 4.2. Schematic structure of the water boxes of the entire model showing routes of water exchange (arrows) between adjacent boxes.

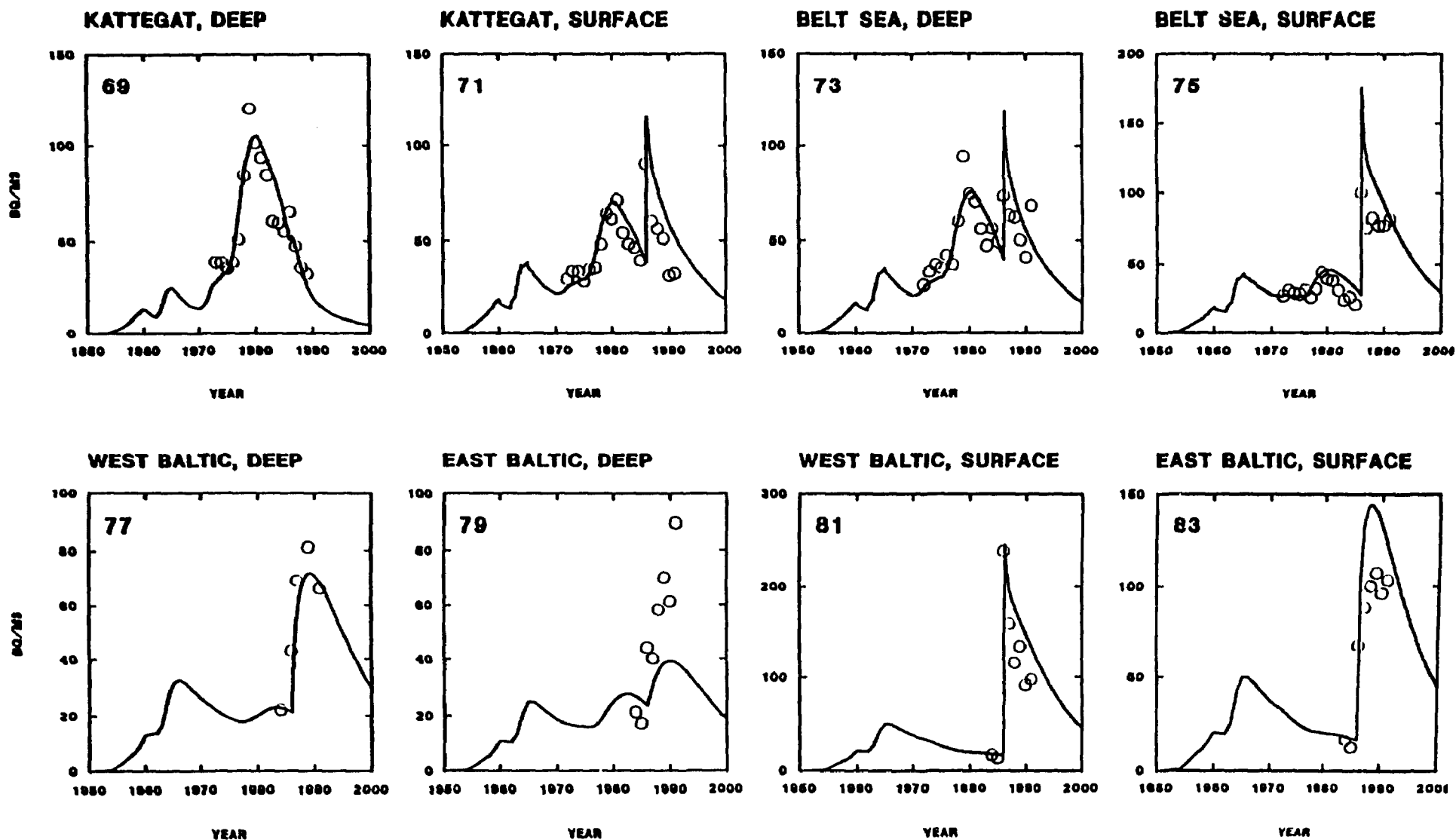


Figure 5.1.1. Calculated and observed concentrations of Cs-137 in seawater (Bq m<sup>-3</sup>) in the water regions of the Baltic Sea. The full lines show the predicted levels and the circles show the annual arithmetic mean values of the observed levels.



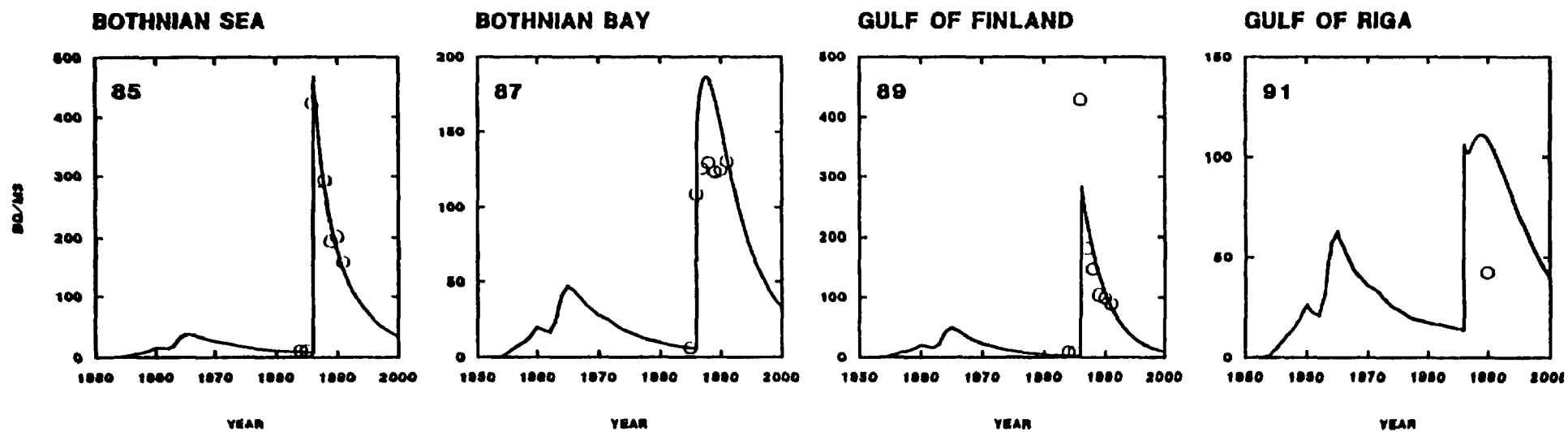
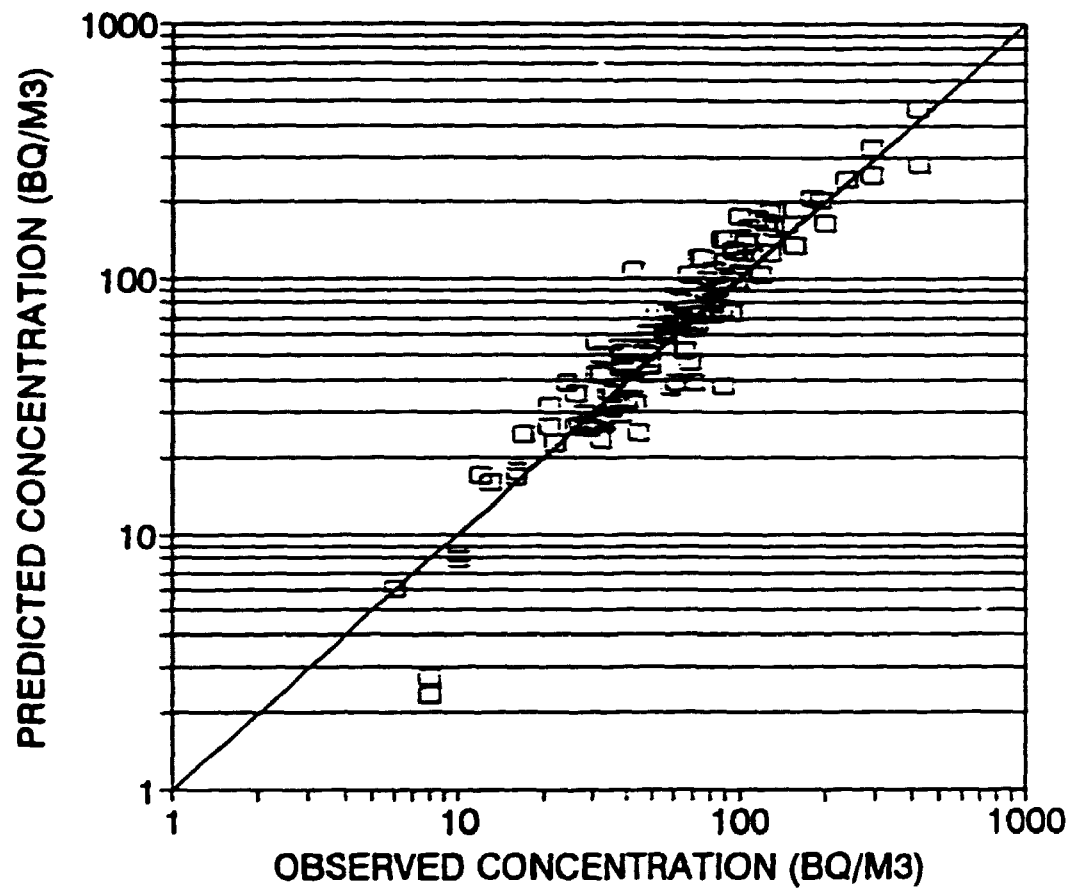


Figure 5.1.1. Continued.

# CS-137 IN SEAWATER



*Figure 5.1.2. Scatterplot of predicted and observed annual mean concentrations of Cs-137 in Baltic seawater. The line indicates 1:1 relationship.*

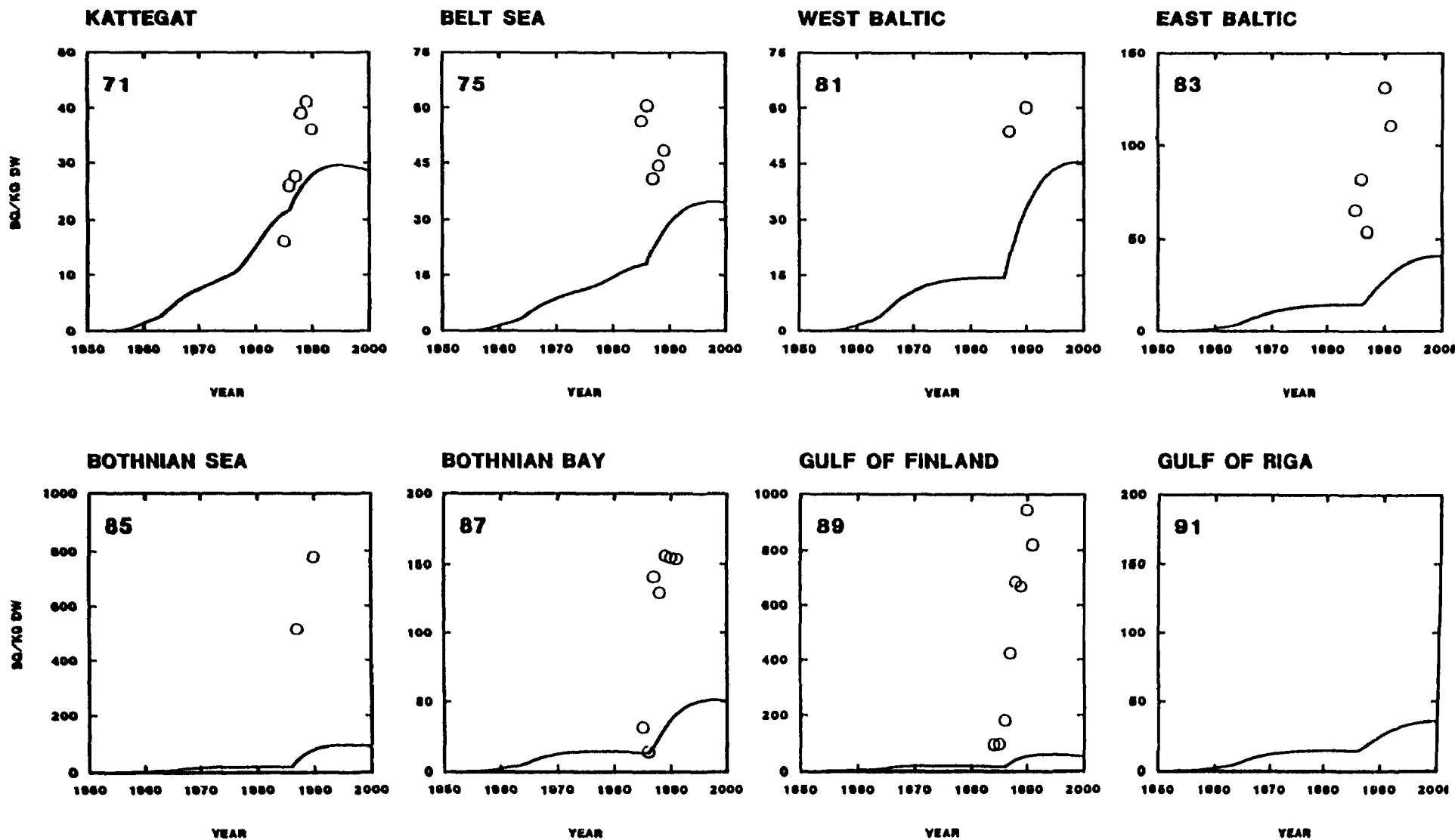
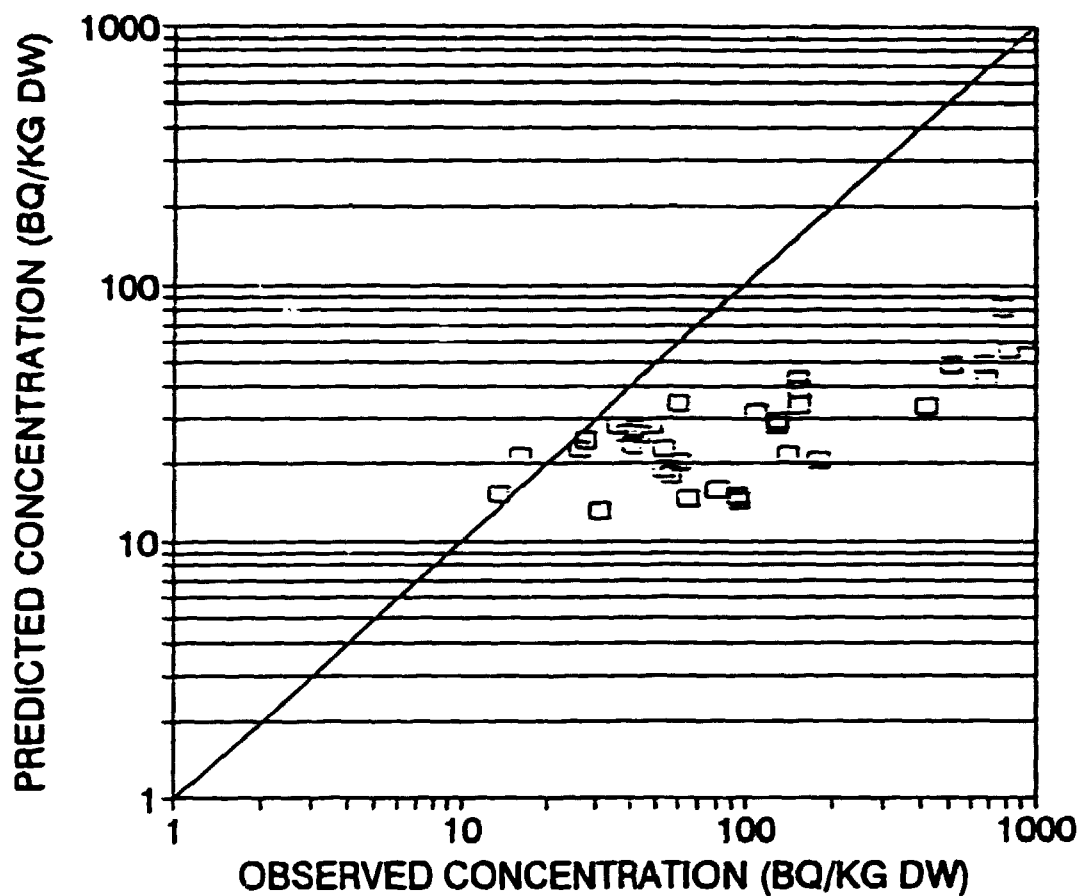


Figure 5.1.3. Calculated and observed concentrations of Cs-137 in top sediment ( $Bq\ kg^{-1}\ dw$ ) in the regions of the Baltic Sea. The full lines show the predicted levels and the circles show the annual arithmetic mean values of the observed levels.

# CS-137 IN SURFACE SEDIMENTS



*Figure 5.1.4. Scatterplot of predicted and observed annual mean concentrations of Cs-137 in surface sediments. The line indicates 1:1 relationship.*

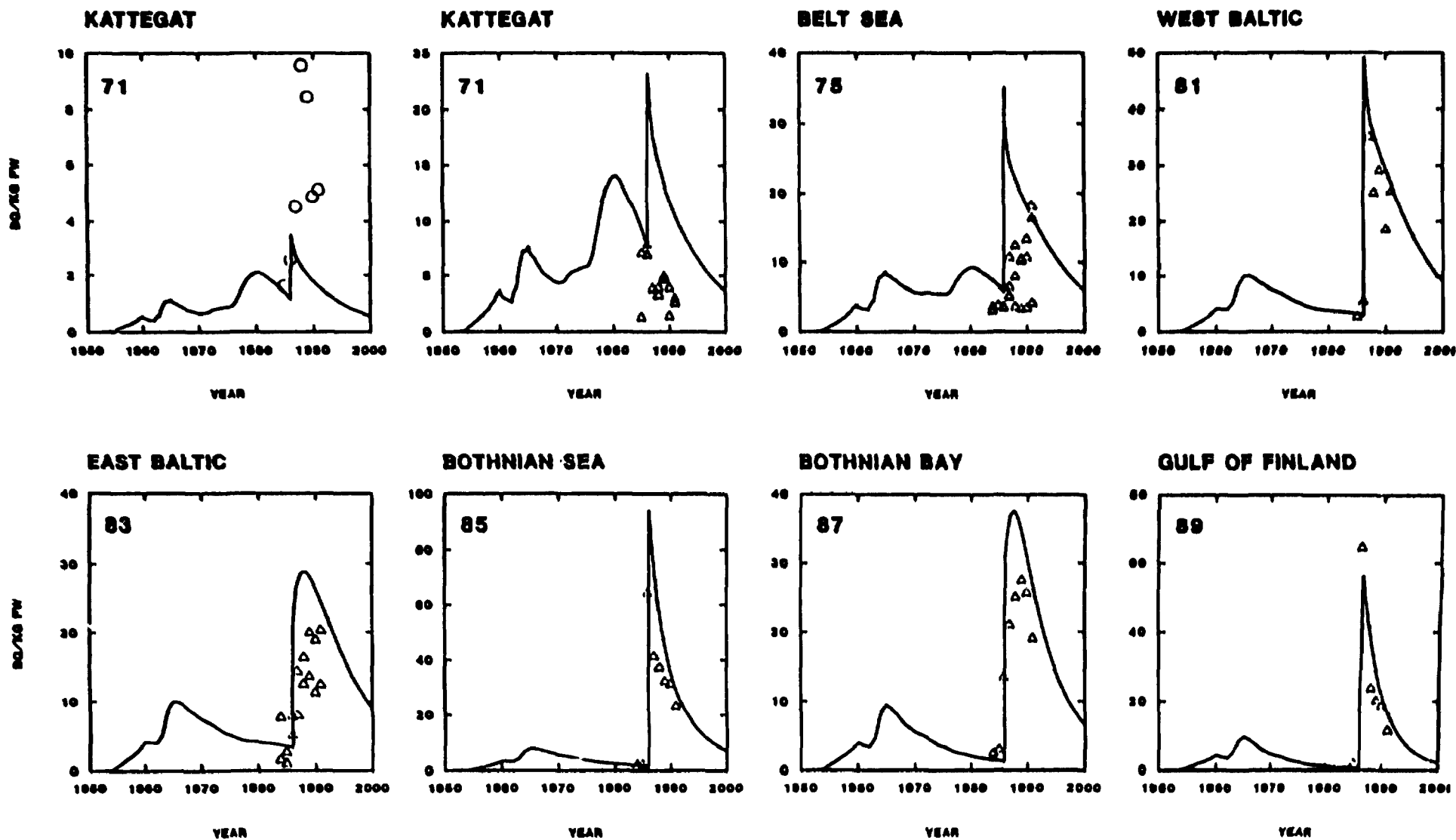


Figure 5.1.5. Calculated and observed concentrations of Cs-137 in biota (Bq kg<sup>-1</sup> fw) in the regions of the Baltic Sea. The full lines show the predicted levels and the circles and triangles show the annual arithmetic mean values of the observed levels in molluscs and fish, respectively.

# CS-137 IN BIOTA

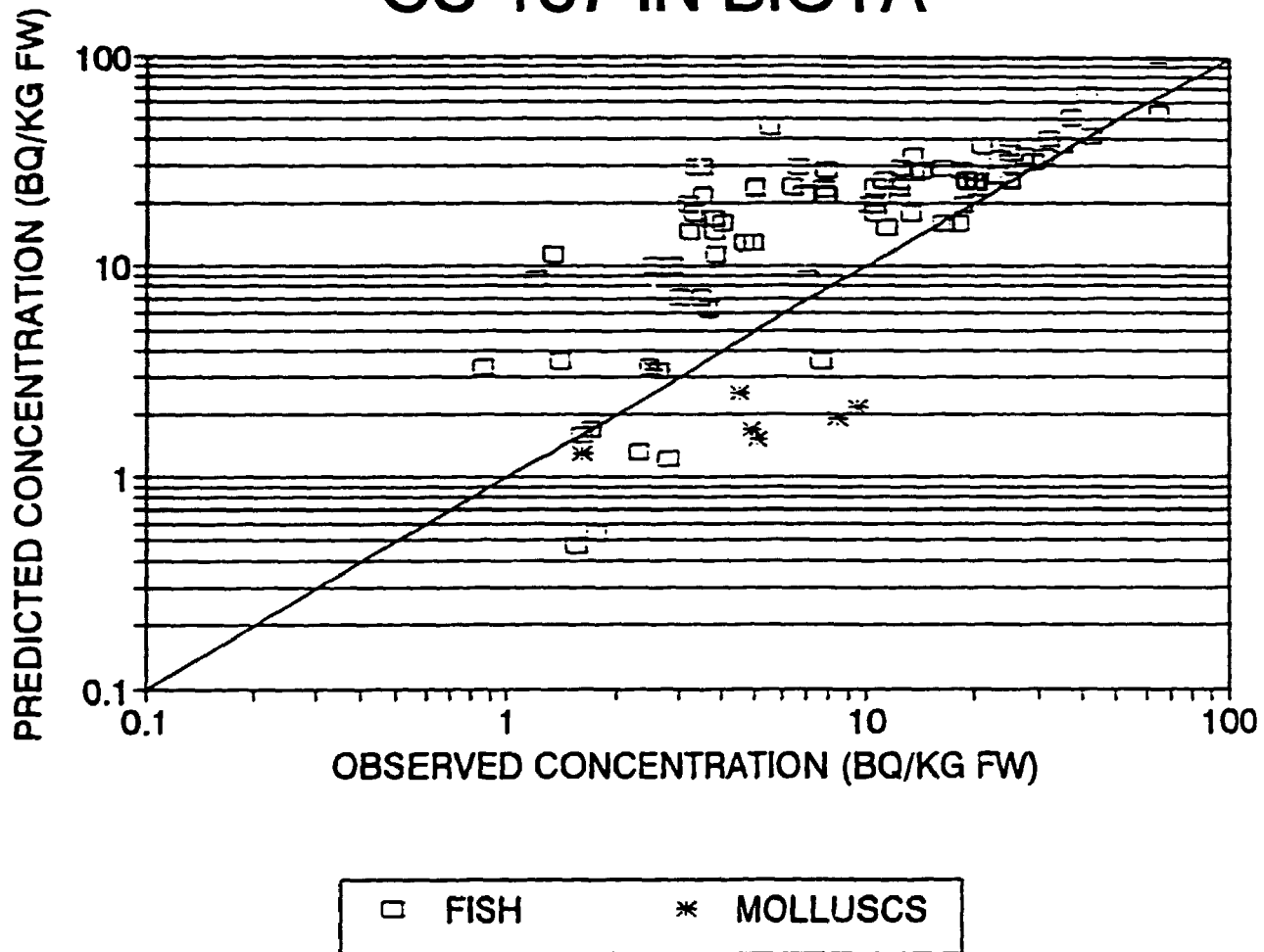


Figure 5.1.6. Scatterplot of predicted and observed annual mean concentrations of Cs-137 in biota. The line indicates 1:1 relationship.

# CS-137 INVENTORY IN THE BALTIC SEA

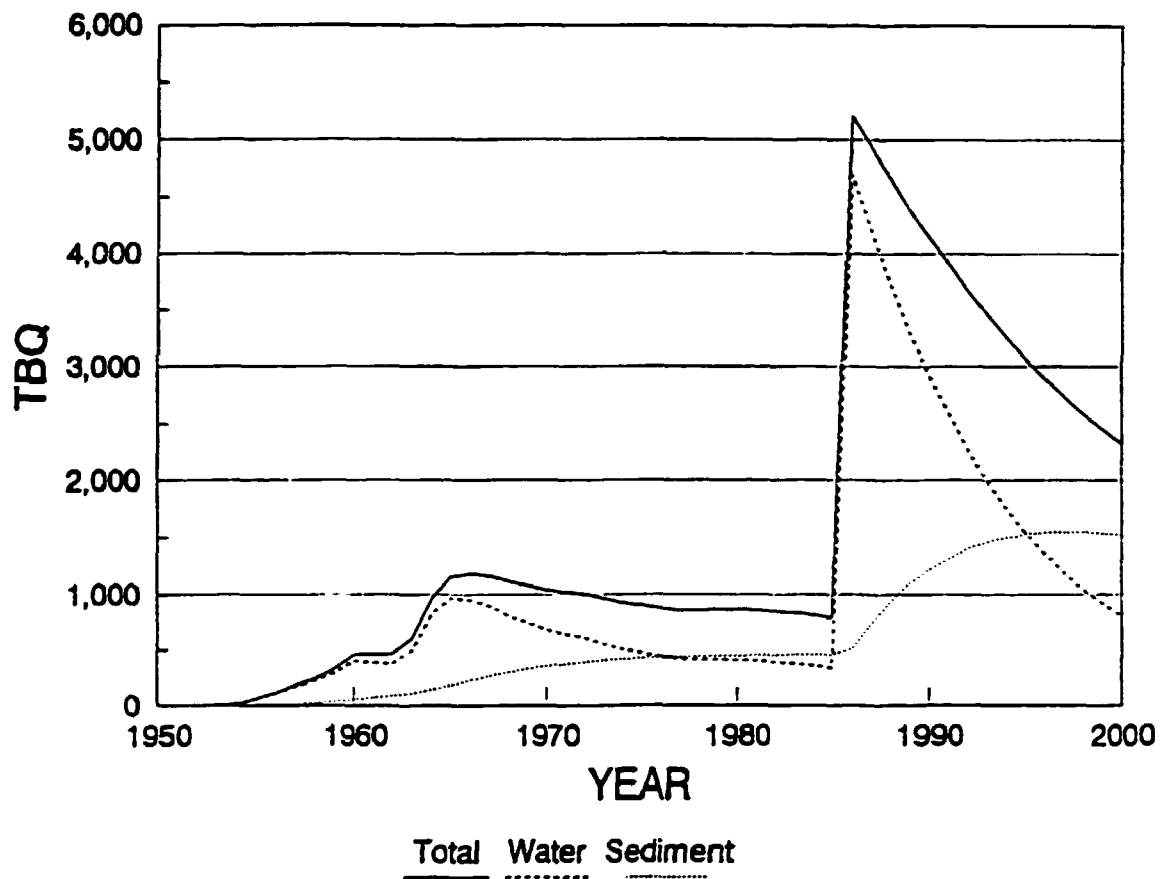


Figure 5.1.7. Predicted inventory of Cs-137 (TBq) in the Baltic Sea in water and sediments shown as a function of time.

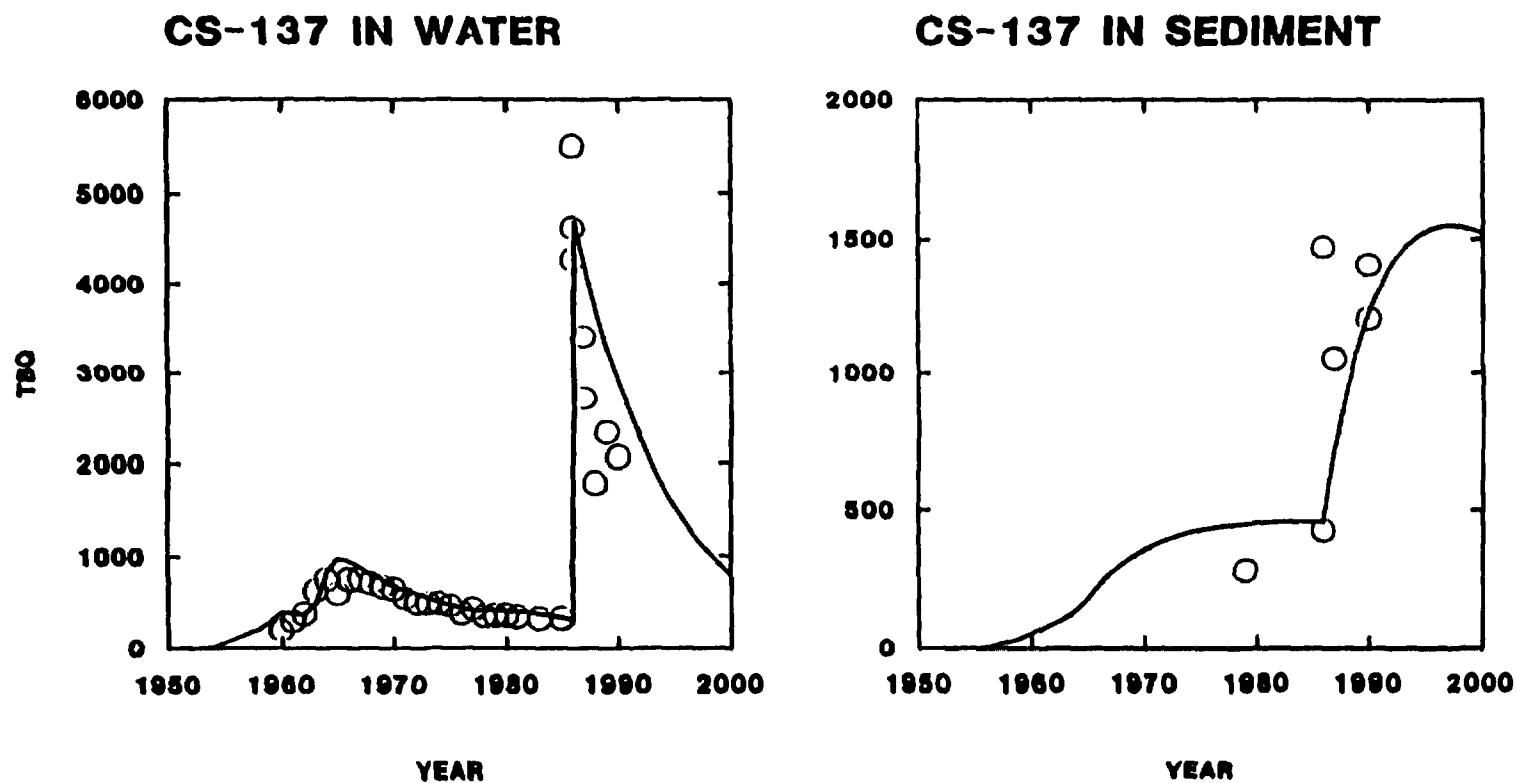


Figure 5.1.8. Comparison of predicted and observed inventories of Cs-137 in Baltic seawater and sediments. The full lines show the predicted levels and the circles show the annual arithmetic mean values of the observed levels.



# CS-137 INVENTORY IN THE BALTIC SEA

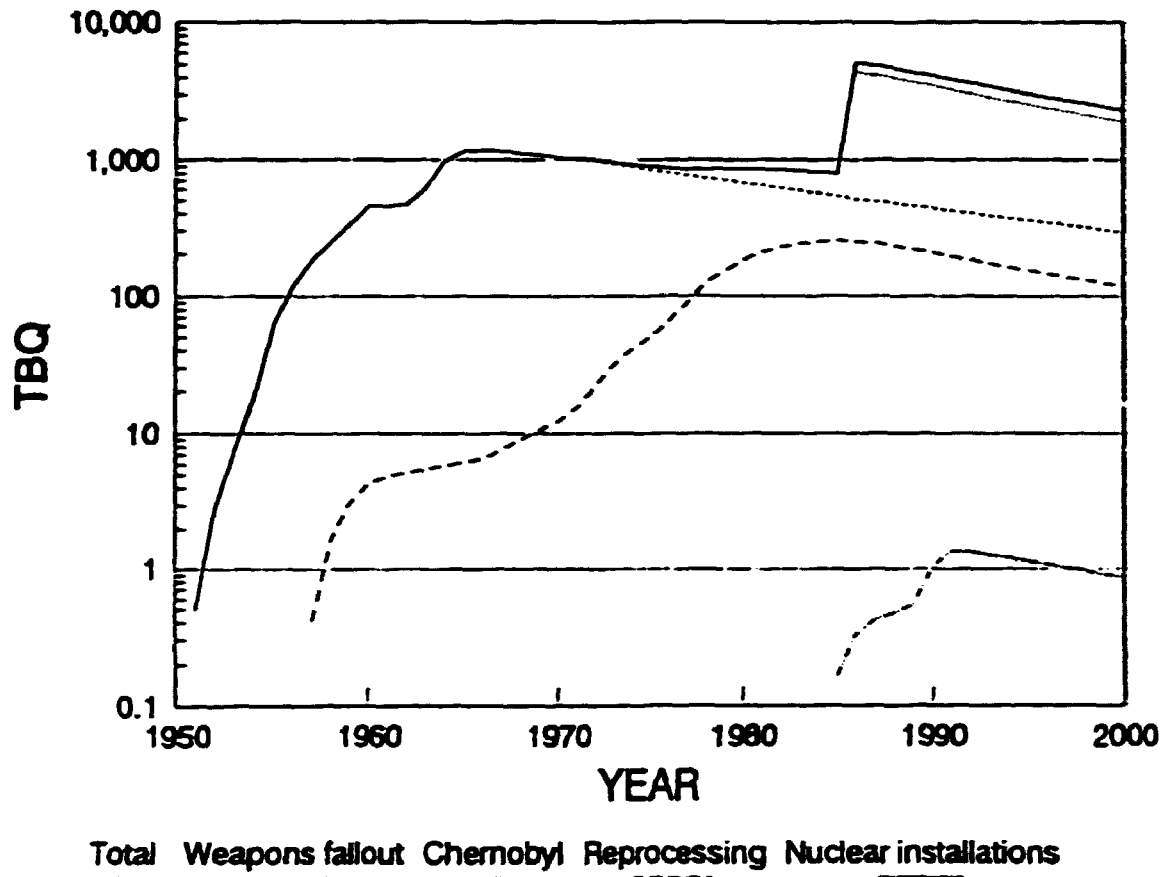


Figure 5.I.9. Predicted inventory of Cs-137 (TBq) in the Baltic Sea broken down according to the sources shown as a function of time.

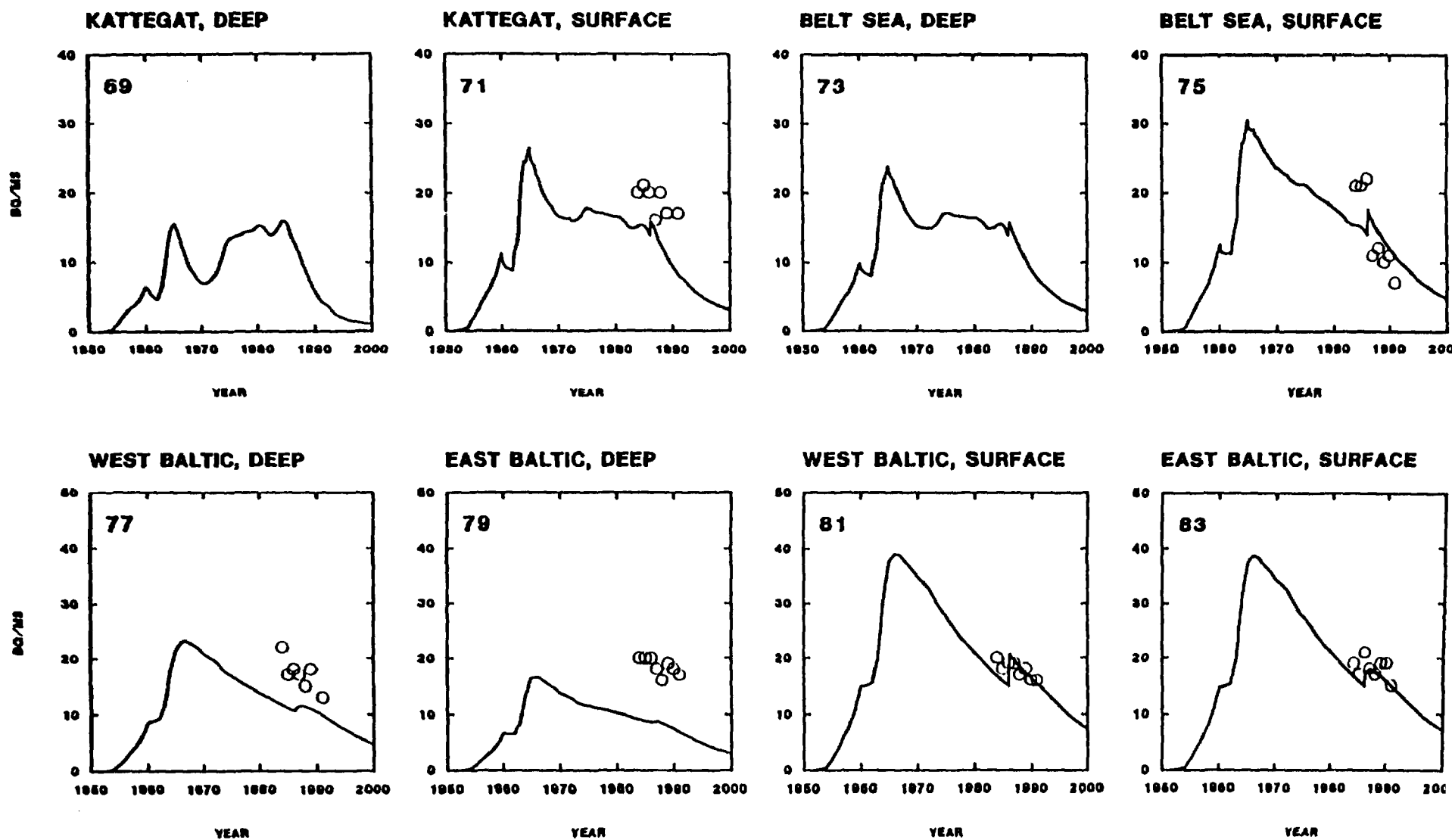


Figure 5.2.1. Calculated and observed concentrations of Sr-90 in seawater (Bq m<sup>-3</sup>) in the water regions of the Baltic Sea. The full lines show the predicted levels and the circles show the annual arithmetic mean values of the observed levels.

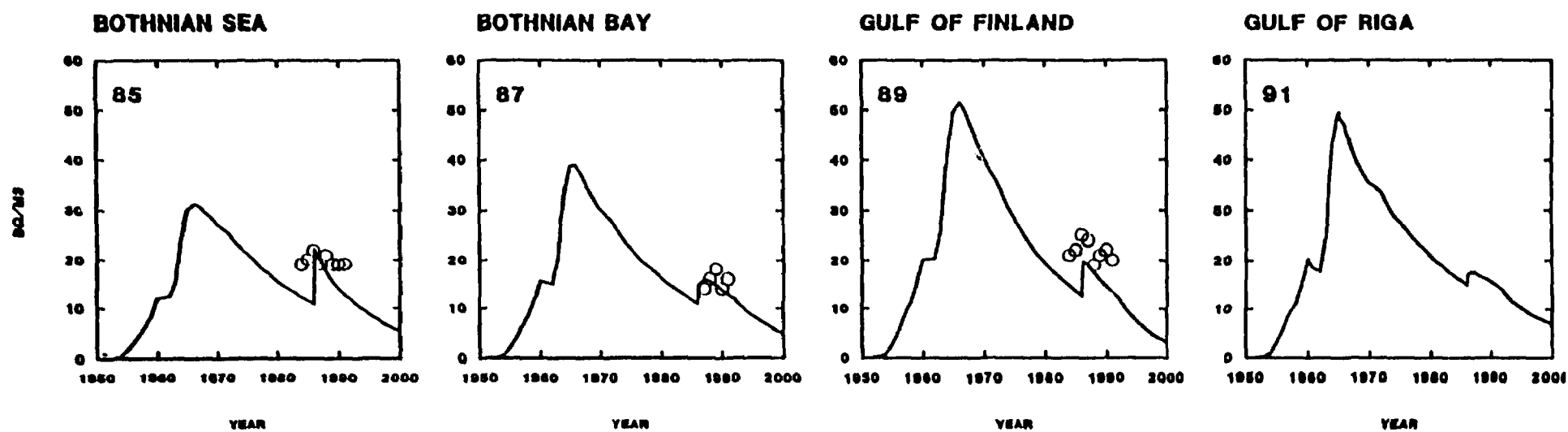
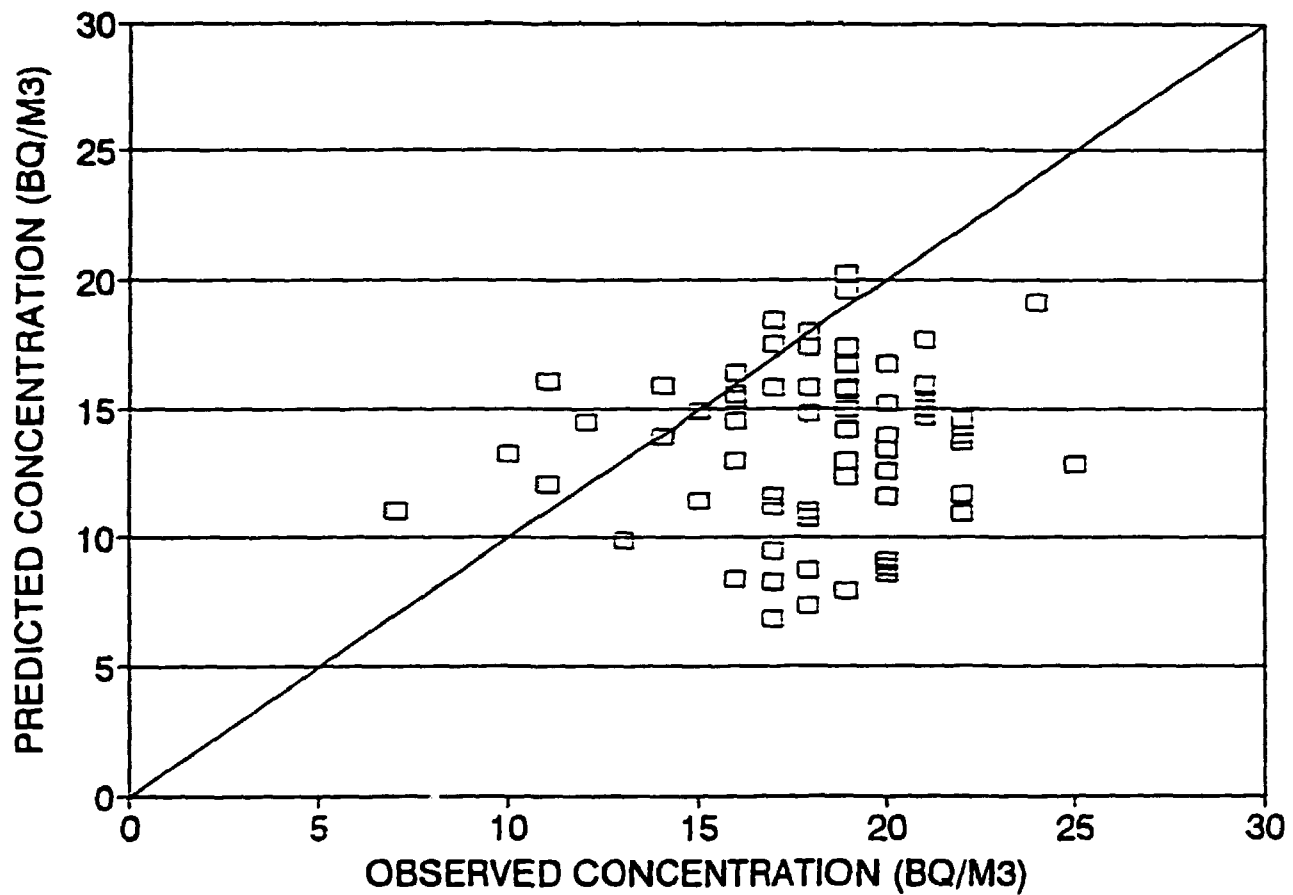


Figure 5.2.1. Continued.

# SR-90 IN SEAWATER



*Figure 5.2.2. Scatterplot of predicted and observed annual mean concentrations of Sr-90 in Baltic seawater. The line indicates 1:1 relationship.*

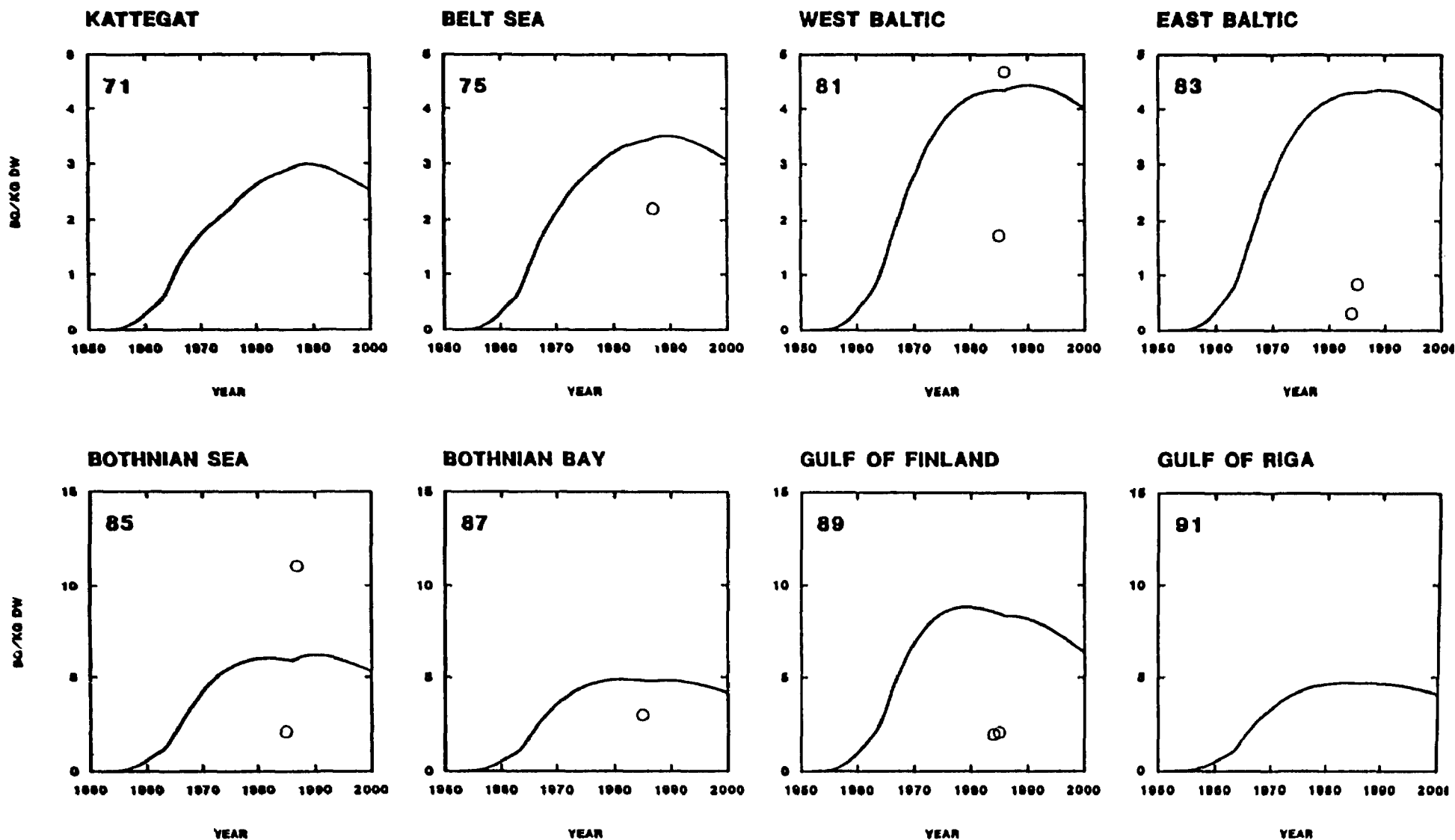


Figure 5.2.3. Calculated and observed concentrations of Sr-90 in top sediment ( $Bq\ kg^{-1}\ dw$ ) in the regions of the Baltic Sea. The full lines show the predicted levels and the circles show the annual arithmetic mean values of the observed levels.

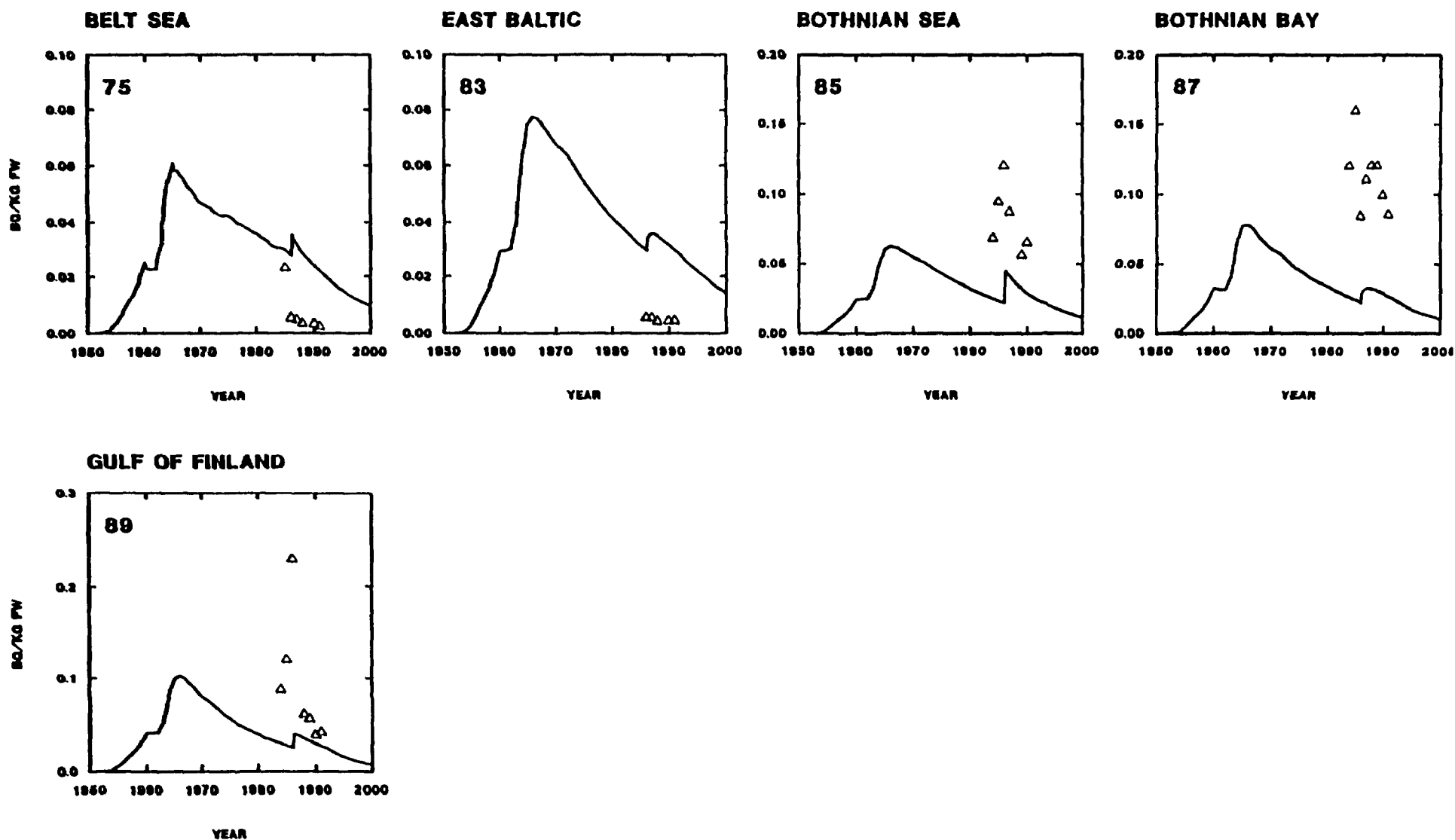
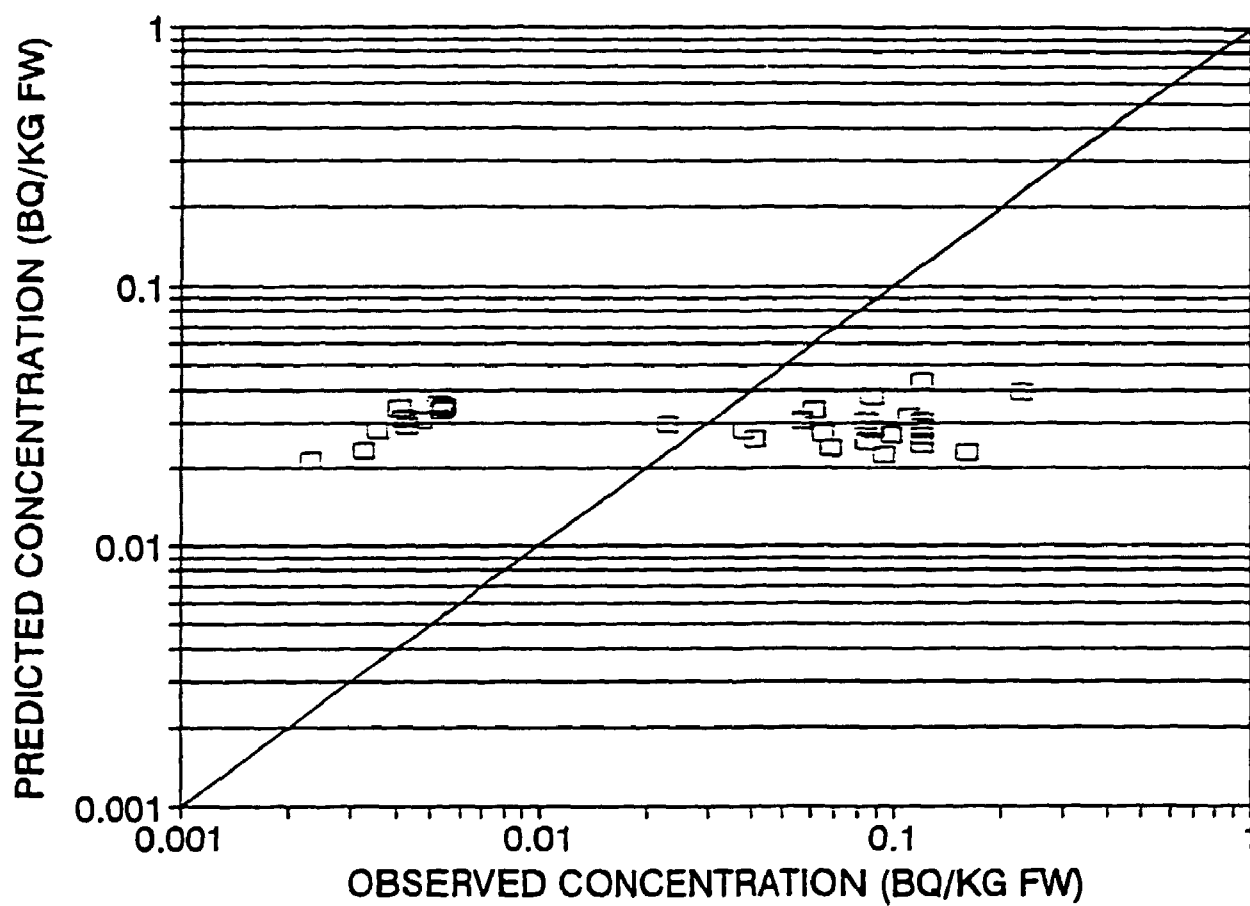


Figure 5.2.4. Calculated and observed concentrations of Sr-90 in fish (Bq kg<sup>-1</sup> fw) in the regions of the Baltic Sea. The full lines show the predicted levels and the triangles show the annual arithmetic mean values of the observed levels.

# SR-90 IN FISH



*Figure 5.2.5. Scatterplot of predicted and observed annual mean concentrations of Sr-90 in fish. The line indicates 1:1 relationship.*

# SR-90 INVENTORY IN THE BALTIC SEA

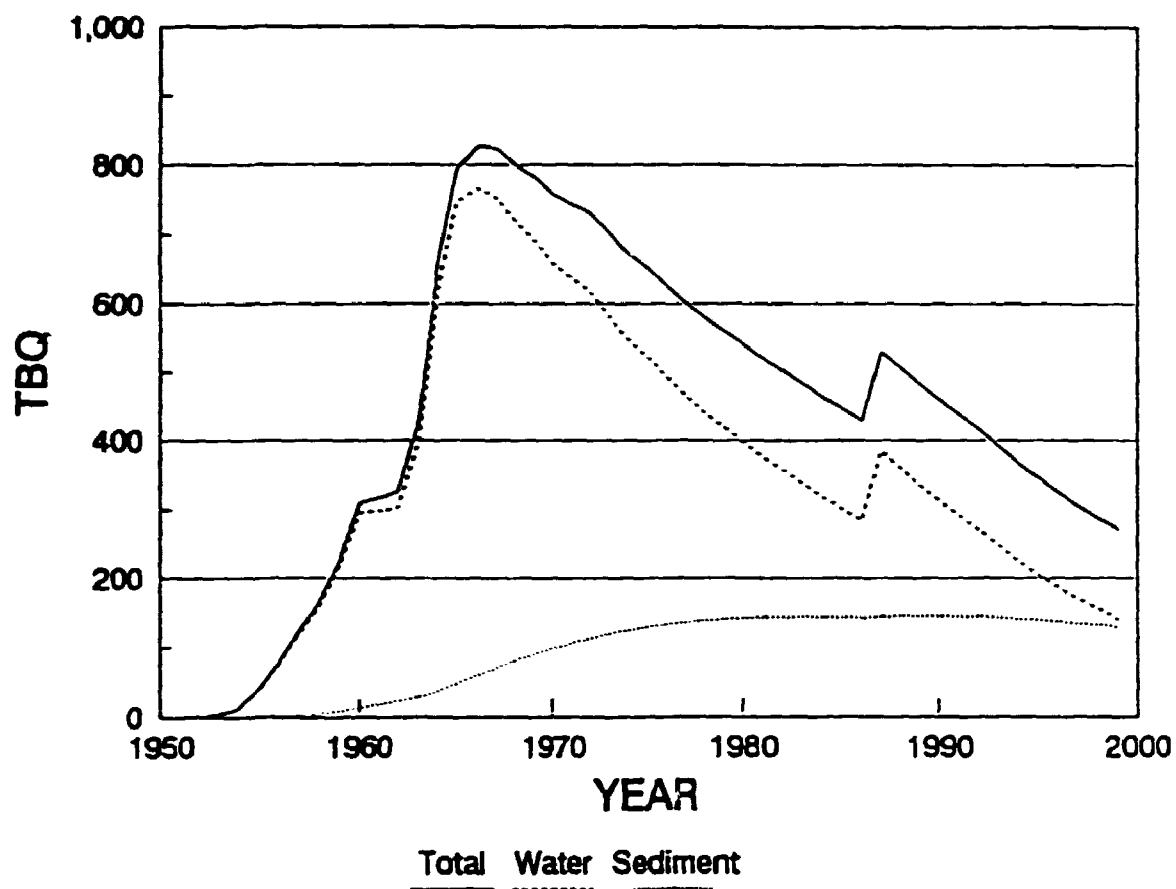
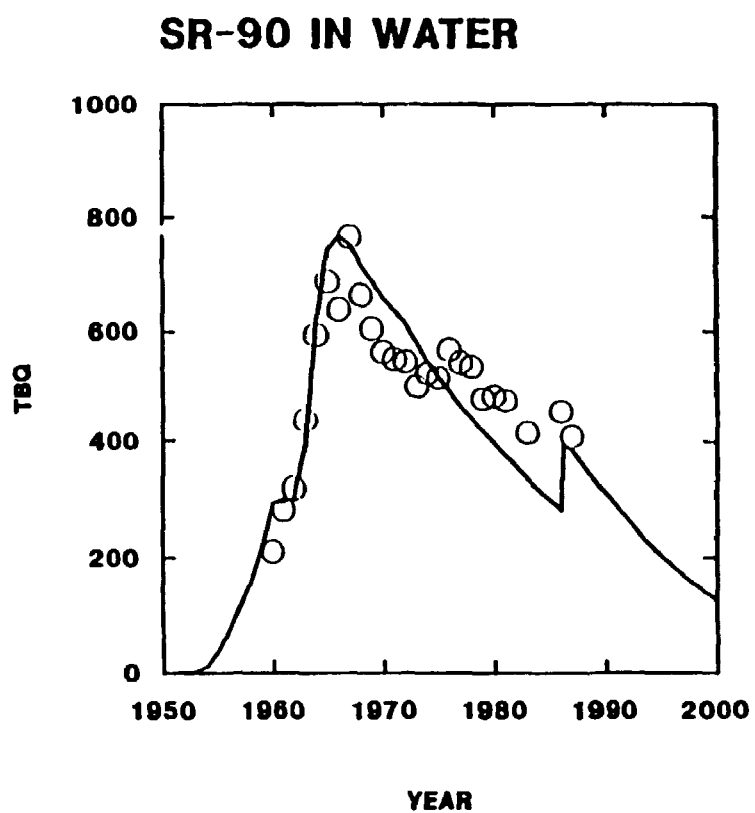


Figure 5.2.6. Predicted inventory of Sr-90 (TBq) in the Baltic Sea in water and sediments shown as a function of time.





*Figure 5.2.7. Comparison of predicted and observed inventories of Cs-137 in Baltic seawater. The full lines show the predicted levels and the circles show the annual arithmetic mean values of the observed levels.*

# SR-90 INVENTORY IN THE BALTIC SEA

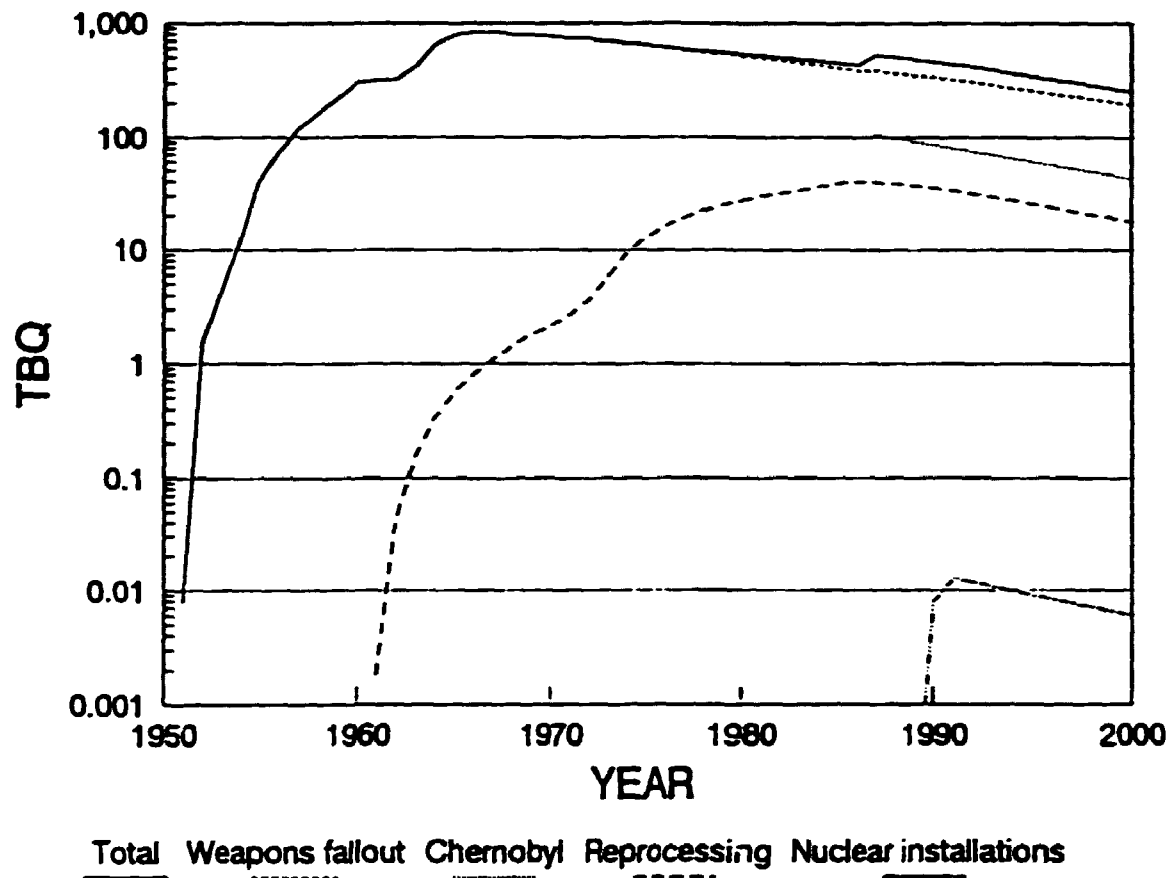
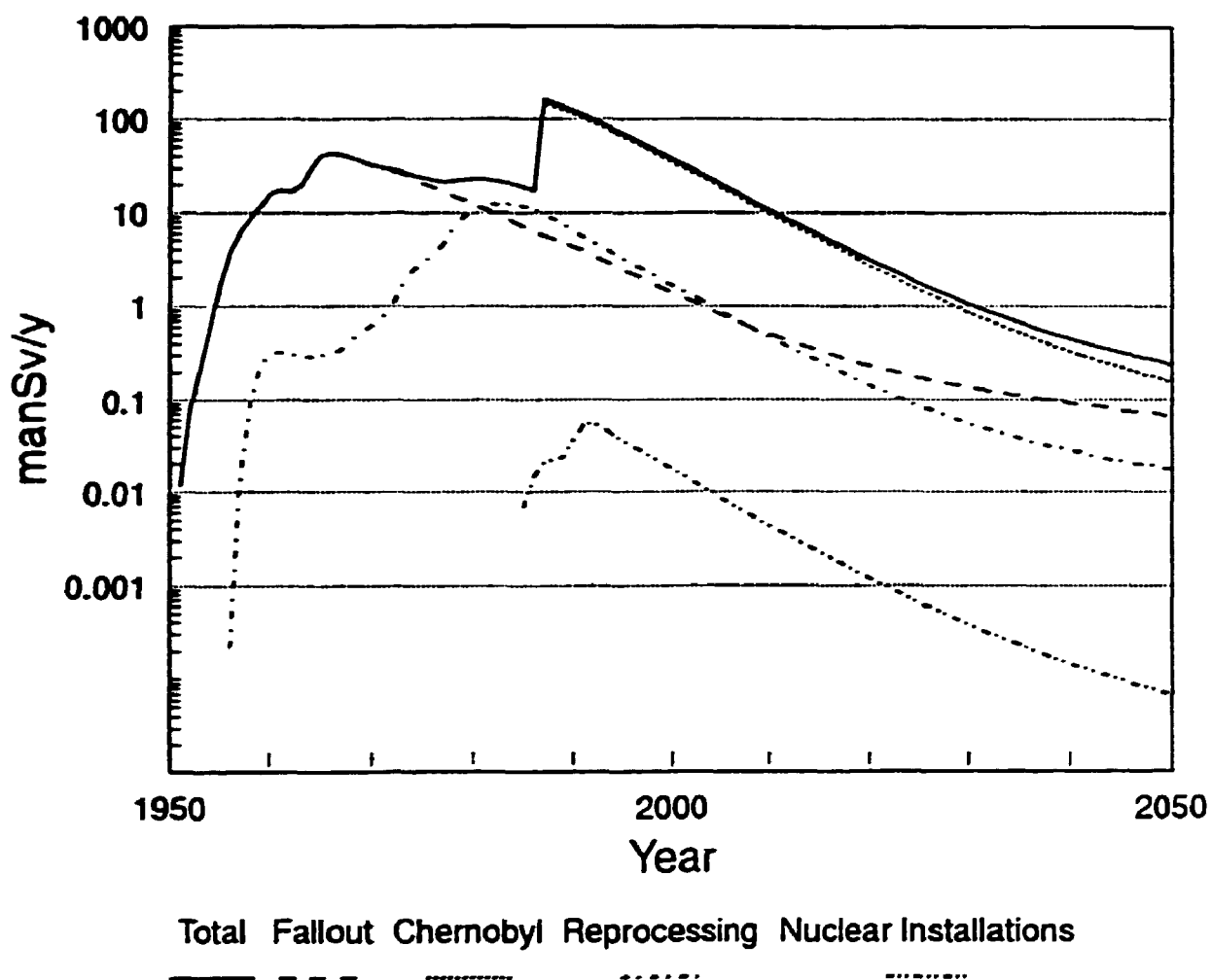


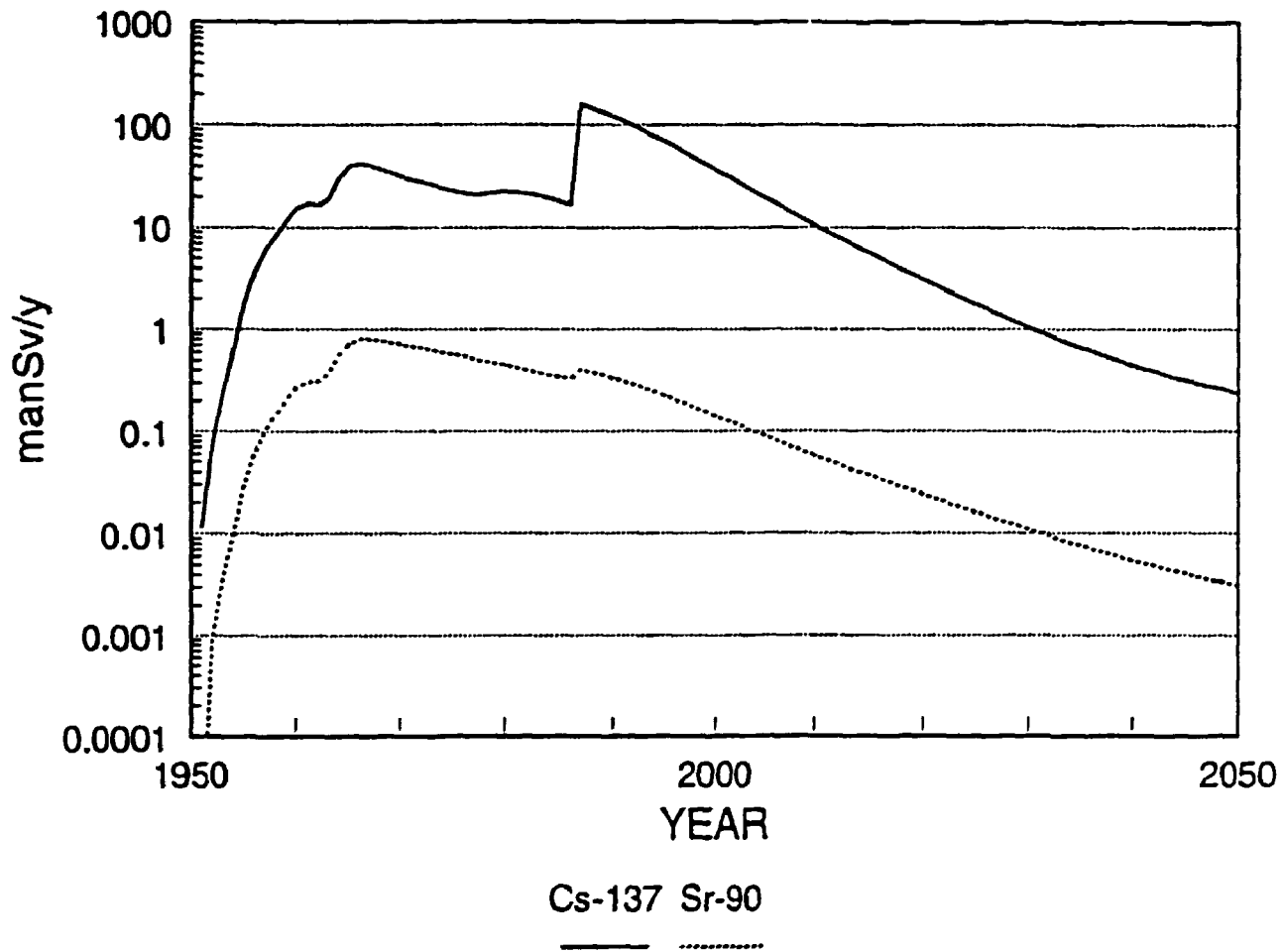
Figure 5.2.8. Predicted inventory of Sr-90 (TBq) in the Baltic Sea broken down according to the sources shown as a function of time.

# COLLECTIVE DOSE RATES



*Fig. 6.2.1. Collective dose rates (manSv y<sup>-1</sup>) to members of the public from the ingestion of Cs-137 and Sr-90 in seafood from the Baltic Sea area according to sources.*

# COLLECTIVE DOSE RATES



*Fig. 6.2.2. Collective dose rates (manSv y<sup>-1</sup>) to members of the public from the ingestion of Cs-137 and Sr-90 in seafood from the Baltic Sea area.*

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**Title and authors(s)**

**The Radiological Exposure of Man from Ingestion of Cs-137 and Sr-90 in Seafood from the Baltic Sea**

**Pilot Project: Marina-Balt**

**Sven P. Nielsen, Mette Øhlenschläger and Olof Karlberg**

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**ISBN**

**87-550-2072-0**

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**ISSN**

**0106-2840**

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**Dept. or group**

**Environmental Technology and Science**

---

**Date**

**April 1995**

---

**Groups own reg. number(s)**

---

**Project/contract no.(s)**

---

**Pages**

**54**

---

**Tables**

**11**

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**Illustrations**

**28**

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**References**

**17**

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**Abstract (Max. 2000 characters)**

This report describes a limited radiological assessment of the collective doses to man from the intake of seafood from the Baltic Sea contaminated with the radionuclides Cs-137 and Sr-90. Information on fisheries statistics is presented. The most important source terms to radioactive contamination of Cs-137 and Sr-90 in the Baltic Sea are identified and quantified. A compartment model for the dispersion of radionuclides in European coastal waters including the Baltic Sea is described and tested by comparing model predictions with observations. Collective doses are calculated with the model for each of the source-term categories.

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**Descriptors INIS/EDB**

**BALTIC SEA; BOX MODELS; CESIUM 137; CONTAMINATION; DISPERSIONS; DOSE RATES; INTAKE; MAN; RADIOACTIVITY; RADIOISOTOPES; SEAFOOD; STRONTIUM 90**

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**Risø-R-819(EN)**  
**ISBN 87-550-2072-0**  
**ISSN 0106-2840**

Available on request from:  
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